ISOTOPES AND RADIATION TECHNOLOGY

A Quarterly Technical Progress Review

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FEATURE ARTICLE:

Particle Accelerator Uses by Industry

TECHNICAL PROGRESS REVIEWS

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Isotopes and Radiation Technology is a Technical Progress Review prepared by the Isotopes Development Center, Oak Ridge National Laboratory, at the request of the Divisions of Technical Information and Isotopes Development, U. S. Atomic Energy Commission. This Review is intended to assist those interested in keeping abreast of significant developments in the fields of isotopes and radiation technology. It is not a comprehensive review of all literature published in this field during a given quarter; rather, it is a mechanism for presenting concise, selected reviews of information on subjects of prevailing Commission interest as it becomes available.

This Review attempts to relate the published results of research and development sponsored by the Division of Isotopes Development to significant developments in radioisotopes and radiation technology, as reported in the world literature. Coverage includes isotope production and development, isotope technology development, isotope applications technology, process radiation development, and radiation processing of food and medical supplies.

In addition to reviews of current literature and special review articles dealing with specific isotopes, facilities, and applications, this publication occasionally contains feature articles prepared by recognized experts on specific topics of current interest. Critical evaluations and interpretations presented are those of the editors and invited reviewers; therefore readers are encouraged to consult the original references in order to obtain all the background of the work reported and the interpretation of the results given by the original authors.

The subject headings listed below have been adopted to help maintain continuity and order, from one issue to the next, in the material reviewed. All reviews, except Feature Articles, will be arranged under these headings; but any one issue will not necessarily contain all the headings.

Isotope Production and Development	Isotopic Power Development
Isotope Technology Development	Applications in Medicine
Food Irradiation	Safety
Process Radiation Development	Isotopes Around the World
Source Development	Miscellaneous Items of Interest

P. S. Baker, Robert H. Lafferty, Jr., Martha Gerrard, D. A. Fuccillo, R. E. Greene, F. E. McKinney, and Joan E. Carden, *Editors*
A. F. Rupp, *Advisory Editor*

Oak Ridge National Laboratory

FEATURE ARTICLE

Particle Accelerator Uses by Industry

By E. Alfred Burrill*

The 575 charged-particle accelerators in the world, besides those used for clinical therapy, are used for radiography, activation analysis, and radiation processing in industry and research, and for contract services. Their special advantages in various fields are discussed.

Since the mid-1930's — well before the days of the U. S. Atomic Energy Commission (AEC), the nuclear reactor, and large quantities of artificially produced radioactivity — supervoltage particle accelerators have been used to study the effects of nuclear particles and ionizing radiations on materials. From the successful use of radium in clinical therapy and industrial radiography, powerful X-ray generators were developed from several principles of voltage generation in the range above 1 Mev. A natural corollary to these practical uses was the fundamental and objective exploration into the chemical, biological, and nutritional changes produced in a great variety of materials and living systems by these supervoltage X rays and the electrons that produced them.

During these formative years, industry became titillated with the many possible uses for these radiations; and the concept of the radiation sterilization of foods, pharmaceuticals, and medical supplies rapidly attracted interest. The reality of supervoltage accelerators for production-line use in foundries and hospitals lent credence to the possibility of adapting these machines for other types of radiation processing.

Shortly after World War II, the interest in production-line irradiation rose sharply, with the formation of contract radiation facilities and companies for manufacturing accelerators. Many companies invested extensively in equipment, services, and professional talent, which were directed toward the further understanding of radiation effects, radiation facility design, and process costs. The heyday of industrial development occurred in the mid-1950's, when it became fashionable for almost every major chemical company to demonstrate a vigorous interest in radiation processing.

Several roadblocks forestalled the general industrial acceptance of the particle accelerator, despite the keen interest in examining the effects of nuclear particles and ionizing radiations on materials and components. It was difficult to find many processes in which radiation was uniquely valuable. In those instances in which radiation did offer promise, the problems of economics and state of the technology hindered industrial acceptance.

Against many odds, therefore, including the stigmas of reliability and economy (not to speak of the reputed and often debated advantages of radionuclidic sources of ionizing radiation), the accelerator is now coming into its own, in designs that are considerably different from those formerly felt to be desirable and with economics and reliability that now cause industry to pay attention. The continuing progress in radiation chemistry, sterilization, solid-state technology, and food processing has also done much to clear the atmosphere in favor of the accelerator. Today nearly a dozen U. S. companies are manufacturing accelerators (Table 1), and well over 300 kw of accelerator-produced radiation power is being applied to

*High Voltage Engineering Corp., Burlington, Mass.

Table 1 U. S. ACCELERATOR MANUFACTURERS

Manufacturer	Accelerator	Nuclear radiations
Allis-Chalmers Mfg. Co., Milwaukee, Wis.	Betatron	Electrons, X rays
Applied Radiation Corp., Walnut Creek, Calif.	Linear	Electrons, X rays, neutrons
Cyclotron Corp., Berkeley, Calif.	Cyclotron	Ions, neutrons
General Electric Company, Milwaukee, Wis.	Resonant transformer	Electrons, X rays
High Voltage Engineering Corp., Burlington, Mass.	Van de Graaff	Electrons, X rays, ions, neutrons
	Insulating core transformer	Electrons, X rays, ions, neutrons
	Tandem	Ions, heavy ions, neutrons
Kaman Nuclear Corp., Colorado Springs, Colo.	Low voltage	Ions, neutrons
Radiation Dynamics, Inc., Westbury Park, N. Y.	Dynamitron	Electrons, X rays, ions, neutrons
	Dynagen	Neutrons
Technical Measurement Corp., North Haven, Conn.	Low voltage	Ions, Neutrons
Texas Nuclear Corporation, Austin, Tex.	Low voltage	Ions, neutrons
Varian Associates, Palo Alto, Calif.	Linear	Electrons, X rays, neutrons

industrial production lines, many of which are on a multishift basis.

In this article a few of the currently accepted accelerator applications, as well as several developments that can lead to industrial usage, are reviewed. Radiographic inspection, instrumental activation analysis, and radiation processing are three industrial areas in which accelerators have been productively used. The clinical use of supervoltage X-ray generators for cancer therapy could also have been included, but I have chosen to confine my remarks on this use to including them in a tabulation of accelerator distribution.

Table 2 DISTRIBUTION OF ACCELERATORS

Application	United States	Western Europe	Other countries	Total
Clinical therapy	101	104	79	284
Industrial radiography	133	14	18	165
Activation analysis	59	11	7	77
Radiation processing and contract service	34	10	5	49
	327	139	109	575

Throughout the world, as of the end of 1964, there were approximately 575 particle accelerators and supervoltage X-ray generators in clinical and production use (Table 2), excluding research installations but including supervoltage X-ray generators above 1 Mev, neutron-producing particle accelerators above 100 kev, and electron accelerators above 300 kev.

Radiographic Inspection

Before World War II the use of X-ray inspection to determine the soundness of a casting or a weldment was generally confined to X-ray energies below 400 kev and usually well below 250 kev. Encapsulated radium salts were occasionally used for the radiography of thick metal sections, but the total availability of radium was limited to a few tens of grams, and a normal capsule held a few tens of milligrams. Gamma-ray exposures were therefore very time-consuming.

The stringent specifications on ordnance material during World War II forced an advance in X-ray inspection that introduced independently (and for different purposes) the resonant transformer, the Van de Graaff, and the betatron. It then became possible to examine, in production quantities, steel castings and weldments of several inches in cross section and torpedo warheads and mines of over 2 ft in diameter. The U. S. firepower, in the form of shells, gun barrels, mounts, tanks, and even submarine components, could now be improved by engineering and production inspection, on the basis of information provided by these new supervoltage X-ray generators.

It is perhaps indicative of things to come that one of the most definitive papers on the characteristics of supervoltage X rays for radiography came from a physics laboratory at the Massachusetts Institute of Technology (MIT), under the supervision of Van de Graaff.[1]

The pragmatic findings of the technician became explainable; the *science* of nondestructive testing was born. This specialized field has been extensively reviewed in the past few years.[2]

It was obvious that these supervoltage radiations should penetrate many inches of steel and 1 or 2 ft of plastic explosive. It was not predictable that the radiographs produced by exposure to these penetrating radiations would have an unusual clarity, despite a low image contrast, which rendered visible a considerable amount of detail, representing less than 1% of the total thickness. The reason for this surprising result was that supervoltage X rays tend to scatter and disperse from their original direction to a far less degree than low-voltage X rays. Scattering contributes nothing to the significant image on the radiographic film; it only creates background radiation that blackens the film uniformly.

Another feature of supervoltage radiography was the practicality of a very small focal spot of electrons impinging on the X-ray-producing target, whence come the X rays in strictly radial directions to cast a sharp shadow on the film regardless of the distance of the defect in the object from the film. Since X-ray intensity increases exponentially with impinging electron energy, the power needed for a particular intensity decreases correspondingly. Supervoltage X-ray targets were therefore required to withstand far less power than their low-voltage counterparts for radiographic purposes, and the electron beams could therefore be more sharply concentrated. Radiographic definition is of great importance, especially in the inspection of thick objects.[3] The Van de Graaff and the betatron provided such sharp images. The resonant transformer compensated somewhat for its larger spot size by combining larger distance with the use of greater X-ray power.

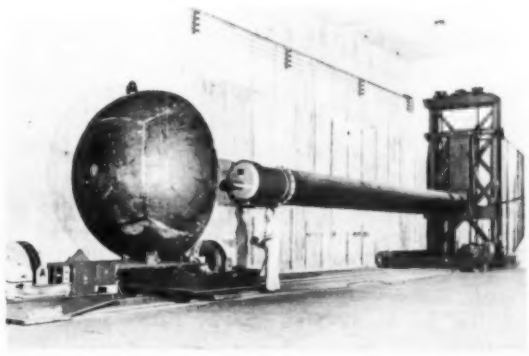
Heavy Steel Industry

The casting or welding of steel (and other metals, such as brass, bronze, and Stellite) has always been accompanied by an uncertainty about the integrity of the finished product due to variations in cooling of the casting and resulting stresses of the metal. Cracks, occlusions of foreign materials, gas pockets, and lack of weld penetration are among the deficiencies to be found in these methods of fabrication. Of all the nondestructive-testing methods, radiography has been most universally accepted because it provides a visual image that can be interpreted on the spot and can subsequently be filed as an indisputable record. The type of defect can guide the engineer to a revision of the casting technique.

As requirements for pressure vessels and for high-pressure valves and components have been increased by the electric-power, nuclear, and ship industries, steel-wall thicknesses have been correspondingly increased to contend with the pressures and temperatures. Normal radiographic practices have thus been extended into the supervoltage, highly penetrating realm as the state of the accelerator art has permitted.

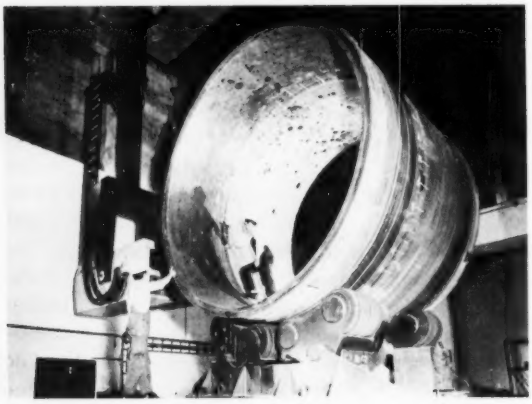
The resonant transformer has for many years been considered the workhorse in supervoltage radiography in the heavy steel industry. Several scores of 1- and 2-Mev X-ray generators of this design are in operation, predominantly in the United States but also in a few installations overseas.[4] Most heavy steel-casting and weldment fabrication is in the range up to 8 in., an ideal thickness for the resonant transformer. This, incidentally, is the realm of the cobalt isotope, whose gamma radiation approximates in penetrating power the X rays from 2-Mev X-ray machines.

The Van de Graaff has contended for the same steel-penetrating region, competitively with the resonant transformer, in a 1- and 2-Mev version. The intensity of X rays from the Van de Graaff has been considerably lower than that from the resonant transformer, but the X rays have come from a 1-mm focal spot—a distinct advantage for precision radiographic work, especially under confining radiographic conditions. Consequently the Van de Graaff has enjoyed a very specialized popularity in the heavy steel industry.[5]



The betatron, with X-ray energies of 10 to 25 Mev, was used during World War II and thereafter[6] and found its place in the radiography of heavy steel castings up to 15 in. thick. The X-ray outputs of these three types of electron accelerators ranged to 200 esu/(cm^3)(min) at a distance of 1 m.

A postwar development, the electron linear accelerator, has changed the complexion and opportunities in supervoltage and high-energy radiography. The Linac principles of operation provide extremely high X-ray intensities at energies above a few million electron volts and, preferably, a few tens of millions of electron volts. Advances in Linac technology have made it possible to mount this device flexibly, to perform production-line examinations of steel thicknesses over 18 in., and to explore experimentally up to perhaps 20 in. Nuclear power components require these massive steel thicknesses with a high degree of integrity. The Linac can provide the source of X rays for such meticulous production inspection.[7]



Inspection of Solid Propellant for Missiles

Radiographic inspection is the only practical method of nondestructive testing that has yet been devised for the inspection of solid-propellant motors for missiles.[8] If there are fissures or inhomogeneities in the rubbery plastic propellant, large chunks can fall away during flight and deflect the missile or vehicle, or excessive burning can occur and cause an explosion.

All types of electron-producing accelerators in the 1- to 20-Mev energy range have been pressed into service for this type of inspection. The betatron was the first logical choice for this application in the earlier days of thick-motor evaluation, but its inherent low X-ray intensity has precluded its production-line use for propellant sections much above 72 in. in diameter. The smaller propellant motors, in the thickness range below 24 in., are being radiographed with Van de Graaff and resonant-transformer X-ray generators. The new super motors with diameters well over 100 in. require not only great X-ray penetration but also high X-ray intensity, so that an exhaustive examination, involving hundreds of square feet of X-ray film, can be done with minimum delay.

The U. S. Air Force now uses 25-Mev electron Linacs whose X-ray output is in the range of 25,000 esu/(cm^3)(min), measured at 1 m from the X-ray target. This is about 200 times more powerful than the output from conventional supervoltage X-ray generators and permits a corresponding reduction in radiographic exposure time.

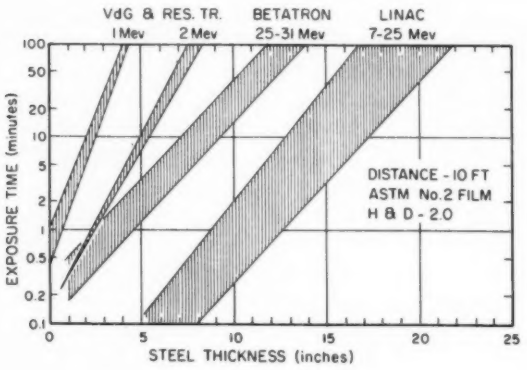
The radiographic requirements of an X-ray generator for solid-propellant inspection include not only sufficient penetration into the material and intensity to accomplish the job as rapidly as possible, but also the finest of image definition and freedom from X-ray scatter that would otherwise obscure the significant X-ray shadow. Small cracks or laminations, perhaps a few thousandths of an inch in cross section, must be discerned through several feet of propellant mass.[3] Changes in homogeneity must be detectable anywhere in the volume. The bond between plastic propellant and motor case must be assured, lest a spurious burning take place at the periphery of the mass rather than out from the center. Supervoltage X rays have the characteristics that are necessary for this type of inspection. In a complete X-ray examination, the volume of the motor is generally evaluated. In addition, tangential shots are often made to check unambiguously the boundary between plastic and casing.[9]

Miscellaneous Radiography

The development of jet aircraft has exploited metallurgical and metal-casting techniques for the blades of the gas turbines that are the driving force of the modern airplanes. Inspection of these blades is vital because one failure could cause the instant demolition of the entire engine and the aircraft. The turbine bucket blades are not thick; in fact, they can be radio-

graphed quite readily with 0.250-Mev X rays. Unfortunately the large crystal structure of the high-temperature nickel or cobalt alloys sometimes creates "ghost images" or diffraction patterns on the radiographic film, which by the uninitiated could be mistaken for cracks or porosity in the blade. Sound blades were often discarded, thus increasing the cost of engine fabrication. Alternatively, defective blades were passed because their defect was unfortunately interpreted as a spurious image. A supervoltage technique was developed, using 1-Mev X rays, in which sufficient freedom from spurious images was obtained but, at the same time, an unambiguous image of the casting was provided.[10]

Some types of nuclear fuel elements are based on strip stock of uranium metal, bonded to a light metal casing for protection and heat conduction. Ingots of uranium are rolled out to the appropriate thickness to form these strips. The integrity of the ingots is therefore important, because any inclusion of foreign matter or gas released by casting ultimately forms an undesirable lamination and contamination in the final rolled product. Uranium is extremely dense and high in atomic weight; it is virtually opaque to conventional X rays in thicknesses much above $1/16$ in. Supervoltage X rays have, however, been successfully applied to ingot thicknesses as great as 4 in. of uranium, although 2 in. is perhaps a practical upper limit for production-line inspection.[11,12] X-ray energies above 3 Mev become less effective for



this type of inspection because of excessive absorption by the production of electron–positron pairs in the material. Hence accelerators in the 2- to 3-Mev class are used for this specialized radiographic work.

Activation Analysis

Neutron activation of materials has long been used as a means of calibrating the particle energies of accelerators and, before the development of instrumental neutron detectors, was one of the few reproducible methods of measuring accelerator-produced neutron outputs.

The use of neutrons specifically for elemental analysis began to attract interest following early work with neutrons produced by cyclotrons[13] and nuclear reactors.[14] Since that time, activation analysis has developed along several lines: (1) using thermal neutrons from reactors, isotope sources, and accelerators; (2) using fast neutrons from accelerators; and (3) using high-velocity charged particles from accelerators.

The concept and detailed use of activation analysis has been covered in previous issues of *Isotopes and Radiation Technology*.[15] This article is therefore devoted to a few of the more interesting and provocative applications of accelerator type neutron sources.

The analytical chemistry laboratory or the production line is not the only logical spot for activation analysis. The oil-exploration and well-surveying groups in the petroleum industry have found this technique to be of some usefulness in the survey of the many strata of the earth's crust down a drilled well.[16] By means of very compact accelerator systems, combined with miniaturized radiation-detection circuits, a log of the constituents in the strata can be continuously recorded as the entire device is lowered down the well. Knowledge of the difference between water deposits and oil-bearing shale can be readily obtained by this method. In fact, the perfection of the oil-well logging method has resulted in the consideration of similar instrumentation for the forthcoming survey of the lunar surface by "soft-landing" spacecraft.[17]

Some very specialized analyses require the discernment of infinitesimal quantities of a constituent (measured in parts per million or even parts per billion). Intense neutron fluxes from reactors are needed for this very sensitive work. The industrial use of activation analysis, on the other hand, exploits the routine assay of minor, and sometimes minute, components in the range down to a few tenths of a percent concentration. Here, the instrumental analytical technique, with neutron-producing

accelerators, is being used because of its rapidity, reproducibility, and adaptability to continuous-flow applications.[18,19]

Steel Industry

The basic oxygen furnace has considerably influenced the steelmaking industry during the past 10 years. It is important, in this process, to make accurate determinations of the oxygen in steel because oxygen-containing compounds precipitated during solidification have important effects on the properties of steel. The development of rapid steelmaking techniques such as the basic oxygen process, along with a demand for "cleaner" steels, has created a need for extremely rapid and reliable oxygen determinations. Activation analysis with fast (14-Mev) neutrons from accelerators operating at 0.15 to 0.40 Mev has provided a rapid method for determining parts-per-million amounts of oxygen in steel. With the neutron technique, large samples (>10 g) can be analyzed accurately in half the time needed for much smaller samples with other techniques. The system is nondestructive, and the same sample can be rerun if necessary. About 2 min is needed to make a complete oxygen determination.[20]

The use of deuterons as an activating agent for carbon determinations in the surfaces of steel has been postulated. At bombarding energies above 0.4 Mev [the threshold for the $^{12}C(d,n)$ reaction], low carbon contents common to steel (0.02%) can be determined by observing the neutron emission from the reaction. Deuteron energies of 1 or 2 Mev provide better neutron yields and, hence, greater sensitivity. Although this technique has not yet been applied for production-line analysis, it indicates the versatility of the activation technique.[21]

Chemical Industry

Several oil companies have adopted activation analysis for use not only in their analytical laboratories but also for process control. The major constituents of petroleum (carbon and hydrogen) are not particularly affected by neutron bombardment from accelerator type neutron sources; hence the temporary radioactivity induced in the minor constituents and contaminants stand out in the analysis, with little interference.

The vanadium content in petroleum must often be carefully appraised. Concentrations of a few parts per million are deleterious to catalytic-cracking operations because the vanadium tends to poison the catalyst. Furthermore, fuel oil with even a trace of vanadium produces a stubborn scale in boilers. By means of thermal-neutron activation, rapid determinations of vanadium in crude-oil samples can be made with sensitivities well below 1 ppm.

Low concentrations of the halogens (fluorine, chlorine, bromine, and iodine) can be readily determined by the neutron technique.[22,23] Nitrogen[24] and oxygen[25,26] contents can also be measured. The importance of the activation method lies in its rapidity and simplicity, because little sample preparation is needed, and in its adaptability to analyses that cannot be obtained by conventional methods.

The assay of certain types of ores and metals for small concentrations of, for example, silicon, aluminum, manganese, magnesium, and beryllium is receiving considerable attention. The use of charged-particle activation is being explored as well as the fast- and thermal-neutron technique. In the charged-particle case, the surfaces of samples are analyzed[27] because of the low penetrating characteristic of the charged particles such as protons, deuterons, or ^{3}He.

Accelerators that produce positive ions in the 2- to 3-Mev range have been used to analyze clays, ores, gases, and metallic surfaces with a sensitive method borrowed from nuclear physics—the observation of the energy of the elastically scattered protons or deuterons from the surface of the sample. Depending on the atomic weight of the struck nucleus, the scattered projectile gives up a certain amount of its energy. The energy spectrum of the scattered particles thus reveals the elemental analysis of the bombarded surface, both qualitatively and quantitatively.[28]

Miscellaneous Applications

It is difficult to draw the line between research and industrial applications of activation analysis, because the technique itself is research-oriented and is constantly being improved and broadened in scope in both laboratory and industry. The increasing variety of accelerators that are becoming available tends to improve the acceptability of the method.

One thing is apparent: activation analysis, whether by neutrons of whatever energy or by charged particles of whatever species, derives

its utility from almost purely physical principles and from continuing explorations in nuclear science by the research physicist. The undesired interference in the scientist's experiment many times becomes the basis for the routine determination of the trace constituent.

The step from activating minor constituents in materials to the production of macroscopic quantities of radioactivity involves a more intense neutron flux (as in the reactor case) or an intense beam of charged particles (the accelerator case). Before the availability of reactor sources of neutrons, cyclotrons were used to produce both neutron-rich isotopes (from neutron bombardment) and neutron-deficient isotopes (from bombardment by protons, deuterons, and helium ions). In many cases the resultant activities were short-lived and had to be produced at laboratories near the ultimate user. Today, of course, reactors are extensively used for the production of neutron-rich isotopes.

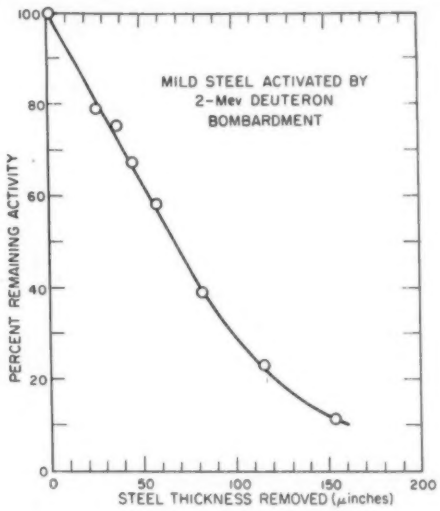
The increasing availability of positive-ion accelerators in universities and industries has led to fields of usefulness that are not, strictly speaking, chemically analytical in result. For example, one chemical company has used a trace of radioactive manganese to observe the rate of mixing of a viscous rubber solution in a stirrer-agitated kettle containing more than 50 gal of rubber dissolved in xylene.

Manganese was chosen because of its short half-life (2.56-hr ^{56}Mn). A total of less than 300 μc of this tracer was used. Such an experiment could not have been conducted by using tracers from a remotely located nuclear reactor because of the rapid decay of the isotope.

By means of a 2-Mev accelerator on site, however, little delay was encountered in putting the isotope to use.[29]

By 2-Mev deuteron bombardment of steel components, a preferential distribution of surface radioactivity can result. The wear of the component in a dynamic system can be observed by monitoring the residual acitivity of the irradiated sample. For lubrication studies this method provides a rapid appraisal of the design and frictional wearing of the moving parts. Removal of a few microinches of material can be detected by the deuteron activation method.[30,31]

A promising new neutron-scattering technique has been explored in which the nondestructive analysis of hydrogen-containing materials can be observed. Hydrogen specifically has an exceedingly high cross section for scattering thermal neutrons. If the transmission of thermal neutrons through a sample is observed, the relative hydrogen content can be obtained because the attenuation of the neutrons is proportional to the hydrogen content. The lower limit of sensitivity for this technique is 0.1% hydrogen with small plutonium–beryllium neutron sources, but it can be proportionally improved with the higher neutron intensities available from particle accelerators. The implications of this technique for the steel industry include the possibility of measuring hydrogen embrittlement in special steels.[32]



Radiation Processing

The concept of introducing ionizing energy into commercial products, as a means of controlling a process other than by heat, pressure, electricity, or chemical catalyst, has been an attractive dream for a host of scientists and technologists ever since the early 1930's. Many of the basic reactions that can be induced by ionizing radiation have been discussed in previous issues of *Isotopes and Radiation Technology*.[33] Reducing these fundamental reactions to commercial practice has been a lengthy and frustrating process, but the field of radiation processing is apparently now starting to open up. The role of the electron accelerator in this progress is discussed in this article, with no attempt to include the many contributions from isotope technology.

Polymerization

Irradiated-polyethylene-insulated wire and cable have been commercially produced for several years, using electron accelerators in the 1- to 2-Mev range, primarily in the United States and England. The main reason for irradiation is to improve the heat resistance of the plastic so that higher currents can be sent through wire of a given diameter or, preferably, that wires of smaller diameters can handle the same current.

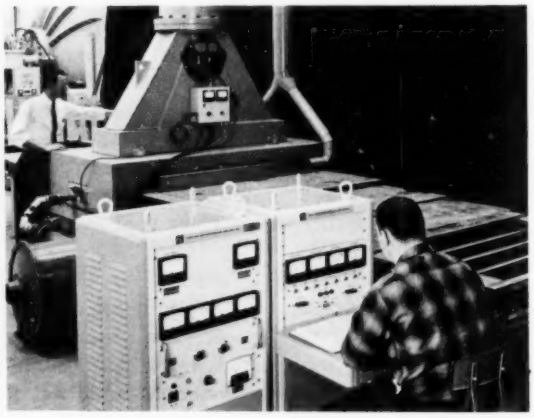
Heat-shrinkable tubing and packaging materials have been produced for several years using accelerators having electron energies as low as 0.5 Mev to penetrate the thin films or tubing walls. The electronics industry has found that heat-shrinkable tubing is invaluable for covering soldered joints or for protecting cable connectors from abrasion or shock. Moisture resistance can also be provided by this general technique, in which the polyethylene tube is slipped over the component of interest and is then shrunk to form a tight fit by the judicious application of heat. At least three processing companies are marketing this type of product, and several powerful electron accelerators in the 0.5- to 3-Mev range are involved in processing the total production of these organizations.

Food is now packaged in heat-shrinkable plastic bags. One company uses a bank of 0.5- to 2-Mev electron accelerators to handle the increasing demand for this packaging material. For example, poultry is placed in an irradiated plastic bag, the air is exhausted, a plastic seal is applied to the opening, and the entire package is then dipped in hot water. The result is a tight, abrasion-resistant package. Furthermore, the removal of the air decreases the opportunity for food spoilage, thus enhancing the storage life of the packaged product.

Miniaturized microwave circuits for missile guidance and advanced communications systems are more practical through the use of solid microwave guide materials that can accommodate several microwave circuits within a few square inches of circuit board. The board is a laminate of polyethylene sandwiched between thin sheets of copper or with copper foil on one side and rigid aluminum sheet on the other. Electron irradiation causes cross-linkages in the polyethylene to improve the heat resistance of the plastic so that it can withstand the intricate fabrication processes that culminate in the final etched circuit.

With electrons of 0.3 to 0.5 Mev energy, it is now possible to cure paint coatings by radiation polymerization, on a variety of flat materials, to yield a finish that is superior, more economical, and quicker to produce than with conventional curing processes, such as infrared drying. A monomer or a prepolymer is applied to the surface of woods, metals, or cellulose products, with no problem of solvent removal, application of heat, or continuing curing beyond the desired degree that results from the use of certain chemical catalysts. The process is virtually instantaneous as opposed to the 20-min drying time that is often encountered in infrared drying ovens. Furthermore, the electron process does not elevate the temperature of the substrate excessively, thereby protecting it against possible undesirable thermal effects such as burning or dehydration. The electric-power consumption of an electron irradiation facility is less than that of a corresponding infrared oven, and the throughput of material is faster with the electron method.[34]

The treatment of monomerically impregnated laminates and veneers by electron beams is a promising new technique for furniture which can permit the use of soft but beautiful types of wood that heretofore have been excluded from



consideration by the industry. Radiation-induced finishes that accentuate the grain of pecan and gumwood also provide high resistance to impact and to staining. The lamination technique is often preferred over the volume-impregnation method (in which the entire substrate must be irradiated by penetrating radiation) because of weight and overall economic considerations.

Other techniques for applying coatings are being investigated, including the graft polymerization (linking a receptive compound to a normally unreceptive polymer) of paper products or textiles, in which the surface of the substrate would thereby accept a more suitable bonding compound or a fast dye or a waterproofing chemical. These processes are in the developmental or pilot-plant stage, but their future possibilities can be readily envisaged.

Ionizing radiation, particularly that obtained from powerful low-energy electron accelerators, has an important role to play in the treatment of surfaces and thin films. Japan has already seen this possibility in the development of their industrial potential since World War II. Their vested interests have not been a deterring factor since most of their factories were destroyed by bombings. Consequently they can be aggressive and imaginative in their efforts to corner a large segment of highly competitive business from the rest of the world. They have selected ionizing radiation from electron accelerators as one of the more important and provocative process tools that provide a different approach and result than would be obtained otherwise. For example, a rigid polyacrylic foam has recently been announced for structural insulation purposes. Electron irradiation is used for the polymerization of this interesting material.

Solid-State Devices

Ionizing radiation can affect solid-state devices, such as transistors and diodes, in a number of ways, depending on the type of radiation, its dosage, and the device in question. In space missions the radiations of outer space tend to degrade the characteristics of solar cells and transistors in the guidance and telemetry circuits. Gross quantities of radiation can wipe out the usefulness of semiconductors in reactor installations. On the other hand, the judicious application of electron radiation in controllable quantities and energies has enabled solid-state device manufacturers to enhance the characteristics of their products in several ways.

The switching characteristics of solid-state diodes for computer applications can be markedly improved by electron irradiation. This improvement is obtainable on a permanent basis for diodes that have silicon as the substrate. Germanium devices do not retain their irradiated characteristics when subjected to increased temperature; the effects apparently anneal out. Typical switching-time improvements for silicon devices are from 10 μsec (unirradiated) to 10 nsec (after irradiation), an improvement of a factor of 10^3. Forward conductance is reduced somewhat, but this is not usually important in computer-switching applications.[35]

Degradation of transistor characteristics would not seem to be an acceptable manufacturing technique. The fact is that it is sometimes more economical to devise an appropri-

ate process for a particular transistor of a very high gain than to adapt the process for a selection of intermediate gains. It thus becomes logical to subject the "standard" transistor to controlled amounts of electron radiation in order to reduce the gain to some predetermined and predictable value.

Other effects of charged-particle irradiation than ionization are now being exploited in the solid-state-device industry. The implantation of heavy ions by particle-acceleration techniques is being used to change passivated silicon substrates to transistors, detector diodes, and solar cells. This new process can have a significant impact on the future of the semiconductor market. Accelerated beams of either boron or phosphorus ions are used to create these devices. The key feature of the process is the ability to implant through the oxide layer of the substrate, thus avoiding the usual problem of surface contamination. This problem has plagued an industry that, up to now, has been utilizing diffusion-doping methods to impart the desired characteristics to semiconductor devices. The ion-implantation method is highly adaptable to integrated circuits because of the possibility of programming the accelerated ions in distribution and in depth, by electronic control.[36]

Sterilization

Many types of products are now being sterilized by ionizing radiations for medicine and industry, to a large degree through the use of electron accelerators.

One of the first production-line electron sterilization applications was at Ethicon, Inc., for the sterilization of catgut sutures. Van de Graaff and linear electron accelerators were installed for research, dosimetry, and actual production use. Ethicon became committed to radiation and adapted a large fraction of their entire production to the radiation process, as a consequence of their pioneering work with accelerators. Ethicon's conversion to gamma-ray sterilization, rather than electron irradiation, is another story discussed in a previous issue of *Isotopes and Radiation Technology*.[37]

The pharmaceutical and hospital-supply industries have examined radiation sterilization in considerable detail, but very few companies utilize this technique in their own plants. In many cases there is not adequate justification for an accelerator because the volume of products to be processed is too small. As a consequence, the contract electron irradiation facility (see discussion under the heading Electron Irradiation Facilities) has served many small needs.

In some cases the electron method does not necessarily produce a unique result; it is merely more economical than alternative sterilizing procedures in which capital investment in equipment would be required.

Disposable plastic blood-donor kits, catheters, and similar hospital supplies are now being sterilized by the radiation technique, generally with electron accelerators. Surgical blades and disposable hypodermic syringes are also being treated by the radiation method. The process is a continuous one, with the devices in their final package, and has few effects on the characteristics of the material being sterilized. Maternity kits, complete with diapers, blankets, masks, and other necessary items, have been experimentally sterilized in the package by electron beams.

Considerable research and a small amount of production-line processing have been done on radiation-sterilized pharmaceuticals. The basic technique has been quite effective and economical. The problem has been to keep pace with the exhaustive testing and clinical documentation, on the one hand, and the obsolescence of the drug, on the other hand. The Upjohn Co., for example, developed a technique of irradiating ophthalmic ointments with 2-Mev electron beams and satisfactorily proved its case to the Food and Drug Administration (FDA), only to find that the ultimate consumer was not particularly interested in the product for reasons not associated with the irradiation technique.[38] The drug industry has also utilized the electron beam from a 2-Mev Van de Graaff for the sterilization of large polyethylene bags used for bulk storage and transportation of antibiotics.[39]

Transplants for the human body have been irradiated and stored very effectually for years. The general method was developed at the Children's Hospital in Boston, using a 3-Mev electron accelerator at MIT, primarily for the sterilization of such vessels as the aorta but has been gradually extended to the sterilization of bone grafts. The materials are usually gathered from human autopsy materials and are immediately frozen to reduce decay. They are then irradiated in the frozen state to render them sterile.

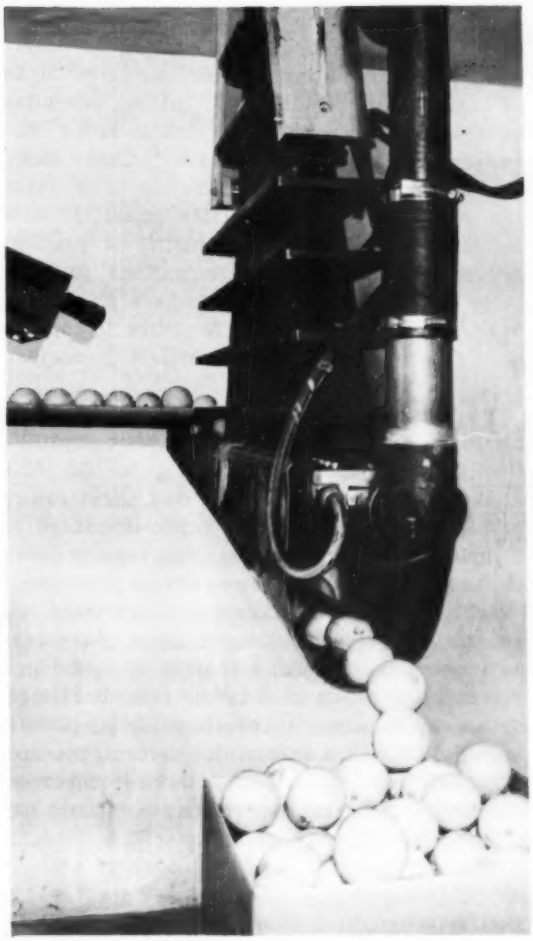
At the New York Orthopaedic Hospital and the U. S. Naval Medical Center, clinical evidence points to the successful grafting of homogeneous bone that had been sterilized with electrons of a few million electron volts energy.[40,41] The effectiveness of the sterilization process is undeniable, but the control of the dose must be precise in order to minimize damage to the bone but at the same time assure complete decontamination. Frozen arterial homografts have been successfully stored and transplanted, using the electron sterilization method with the tissue held at dry-ice temperatures before, during, and after treatment. Among the centers availing themselves of this technique is the St. Louis City Hospital.[42]

Food Processing

The tantalizing prospect of irradiating food to improve its shelf life or to protect the public health—without the application of heat, cold, vacuum, or additives—has been a perennial dream of accelerator manufacturers. A tremendous amount of research has been directed to this prospect over the years, with study of the microbiological and nutritional effects of many types of ionizing radiations on a broad variety of foodstuffs. To date, however, no process has yet been reduced to commercial practice.

Particle accelerators of many types have been used in this extensive and intensive investigation, primarily in university laboratories. The U. S. Army Quartermaster Corps is currently using a 24-Mev electron linear accelerator in its food-sterilization program. Several of the petitions granted by the FDA now include permission to irradiate the particular food with electrons up to 5 Mev energy and, in at least one case, with electrons up to 10 Mev and X rays up to 5 Mev.

Because of the shallow penetrating nature of electrons below 10 Mev, accelerator-oriented research in food irradiation has been primarily directed toward the irradiation of thin packages or layers. Some interesting work has been done on the irradiation of the surface layers of certain foods in which it is desirable to avoid irradiation of the total volume (as in the case of oranges) or in which it is not necessary to process the total volume if the possible infection comes from an invasion through the surface (as in the case of certain carcasses).



Recent experimentation with the electron irradiation of oranges indicates a possible approach to the inhibition of surface and subsurface contamination without affecting the fruit itself. The shallow-penetrating characteristics of 1-Mev electrons are sufficient for providing an inhibiting dose of ionization to the skin. Cobalt gamma rays, on the other hand, are so penetrating that, to reduce subsurface contamination, the entire fruit must be irradiated, thus introducing the possibility of degrading the vitamin C content of the fruit.

Electron Irradiation Facilities

The increasing understanding of the effects of ionizing radiation on materials has created the need for contract radiation facilities. Several accelerator-based centers have sprung up throughout the United States and Western Eu-

rope. These electron irradiation facilities make it possible for companies to explore the possibilities of irradiating specific products, on an electron-beam rental basis, without becoming prematurely involved in the capital outlay necessary for a radiation source.[43] Other companies find that ionizing radiation is of value for some of their specialty products, with insufficient throughput to justify a particle accelerator. Still other organizations, particularly those involved in aerospace programs, avail themselves of these facilities to conduct specialized experiments to simulate the nuclear radiations of outer space.

The character of the business done by these facilities is constantly changing, as companies find their production increasing to the point where they can justify their own accelerator, or as they find that their markets change in complexion either by virtue of increasing competition or by the innovation of new processes.

An example of the latter is a ball-point pen manufacturer who introduced, some years ago, an inexpensive pen with a plastic ink cartridge. The ball was captured in the tip of the cartridge by a swaging process. Unfortunately the plastic material protested against this deformation and ultimately exhibited stress cracking. Electron irradiation of the tip crosslinked the plastic and reduced the tendency to stress-crack. Tens of millions of plastic cartridges were irradiated for this purpose, until the company developed a similarly inexpensive brass-ferrule substitute for the plastic.

These irradiation faciliti .s entertain some fascinating entrepreneurs and inventors — the visionaries of many potentially interesting processes. A few unusual projects have exhibited unqualified technical success but were never adapted to commercial use, for one reason or another. For example, day-old chicks by the dozens have been caponized by discreet applications of 2-Mev electron radiation. The radiation-caponized chicks uniformly grew more rapidly than those that had to recover from their caponizing surgical operation. The irradiation of off-color precious stones by concentrated electron beams has been used to convert what otherwise would have been industrial-grade crystals into beautifully colored gems. Hundreds of stones have been meticulously processed by this technique.

Most accelerator manufacturers maintain irradiation facilities, for either their own development work or demonstration purposes, and occasionally for contract work. The following companies provide contract irradiation on a routine basis:

Electronized Chemicals Corp., Burlington, Mass.
(1-Mev insulating-core transformer, 3-Mev Van de Graaff)

Midwest Irradiation Center, Rockford, Ill.
(10-Mev Linac)

Rad Mat, Gaithersburg, Md.
(3-Mev Dynamitron)

Radiation Polymer Corp., LaGrange, Ill.
(0.5-Mev insulating-core transformer)

Raycon, Windsor, Conn.
(3-Mev Van de Graaff)

Leybold-Hochvakuum-Anlagen (G.m.b.H.), Cologne, Germany
(3-Mev Van de Graaff)

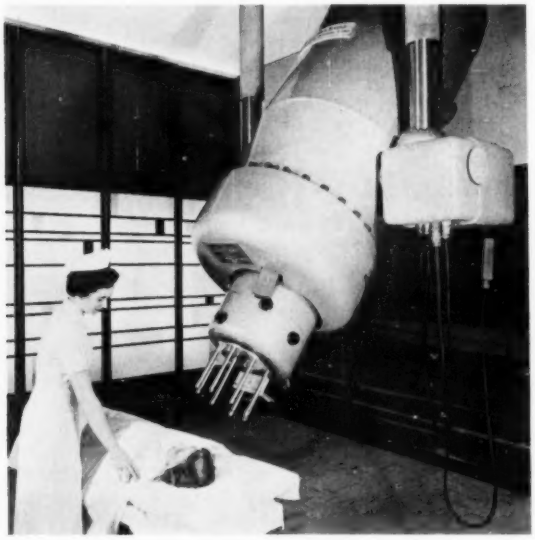
Accelerators or Isotopes?

This article on the industrial uses of radiation would not be complete without some mention of the time-honored controversy concerning which source of ionizing radiation is more effective for the purpose — radioactive isotopes or particle accelerators. Much has been written or spoken on this subject with little coherence or detailed understanding of the industrial problem that might be solved by the use of ionizing radiation. The issue has been cluttered with generalities that have related to the ionizing power available from each type of source; the penetrating characteristics of gamma rays as opposed to the shallow penetration of accelerated electrons; the lack of maintenance required on isotopic sources as contrasted with the continual servicing requirements of electron accelerators — and so forth — not to speak of the exhaustive examinations of the pseudo-economics of both types of sources.

It has been as if all irradiated products were identical; as if all isotope source configurations were standardized; and as if all accelerators were gray. This is indeed not the situation, and comprehension on this subject is starting to be manifest among those (on both sides of the house) who claim to be experts in radiation applications.

In the fields of industrial radiography and radiation therapy, the controversy first started to boil up. Small but intense sources of ^{60}Co with high specific activities (>100 curies/g) were introduced in kilocuries quantities with gamma rays resembling in penetration and in

radiation power the X rays from 2-Mev electron accelerators. The relative economics favored the cobalt sources from the capital-investment standpoint alone. The accelerators still provided more radiation power and considerably greater precision in applying the radiation, by virtue of smaller radiation-source sizes (electron focal spots in the case of the accelerators).



Cobalt sources then started to contribute to the radiation-processing picture, and the "equivalence" of cobalt, X rays, and electrons as ionizing agents became somewhat garbled. For optimum economics, low-specific-activity cobalt (<50 curies/g) is needed, with correspondingly large irradiation areas. The housing for a cobalt irradiation facility has to be at least as large as that for a correspondingly powerful electron accelerator or X-ray generator because of the customarily used low specific activity. For example, a 10-kw electron accelerator of whatever energy has the same total ionizing power as about 0.700 Mc of cobalt. The ionizing power emerges from the accelerator with a uniform distribution up to 6 ft in length and 1 in. in width (0.5 sq ft). A 0.700-Mc cobalt source would present an irradiation area of several square feet. The radiation source must be temporarily removed to a safe position during conveyor servicing. This is usually accomplished by providing a deep water-filled pit into which the cobalt source can be lowered. The water serves as the protective attenuator of the penetrating gamma radiation.

The electron accelerator has also been generalized to excess. From early experience with one or two types of machines, allusions have been made, for all types, to the need for frequent and expensive maintenance. Furthermore, the relatively shallow penetrating nature of accelerated electrons has often been considered to be a general disadvantage to the use of accelerators in processing. The very high ionizing intensity of electron-beam radiation tends to inhibit certain (but not all) chemical processes that proceed quite well with the low dose rate from cobalt. It is often overlooked that electron accelerators can be used to produce intense beams of penetrating X rays, similar in ionizing characteristics to gamma rays.

The facts are now coming to light, and the isotope vs. accelerator controversy is now gradually being replaced by a logical recognition that both types of sources have their places in the scheme of things—whether in radiography, in therapy, or in radiation processing. This logic has come from understanding the details of individual circumstances and industrial requirements.

Each individual process must therefore be specifically appraised, to determine which radiation source is most applicable, from the point of view of penetration (or lack thereof), manipulability, reliability, scale of production, economics, etc. There can be no generalization on this score; there can be only analysis of the specific case. The experience so far in the field has reflected the observation that the source itself is perhaps the least of the concerns of a process; the attending processes, the materials-handling equipment, and the required manpower can easily represent costs that are equivalent to or even in excess of those operating costs for the radiation source and its amortization.

The dog-eared figures of merit are being retired: "capital cost per kilowatt, or per curie." These are being replaced, from process to process, by "cost per pound, or per square foot." Depending on the process, these figures can vary by orders of magnitude—from product to product as well as from source to source.

Both sides in the controversy are now beginning to respect each other in this light. Proponents of all types of radiation sources are now eagerly watching and promoting the field of radiation processing to see in which segment of the overall market each source type can logically fit. Instead of separatism in approach, there is a growing tendency among isotopic and accelerator suppliers toward banding together in solving the common problems of dosimetry, public acceptance of radiation, and radiation standards — with individual applications for radiation to be the challenge for individual source types, in a competitive market.

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Section

I

Isotopes and Radiation Technology

Isotope Production and Development

In this section the second article of a series on the chemical purification of isotopes electromagnetically separated at Oak Ridge National Laboratory is presented. The chemical purification of enriched isotopes of chromium, platinum, and osmium is described. A second paper summarizes the feasibility of recovering transplutonium elements produced in a nuclear explosion (Project Gnome).

The Chemistry of Electromagnetically Separated Isotopes

In this second article of a series reviewing the chemical purification of stable isotopes electromagnetically enriched at Oak Ridge National Laboratory, the purification methods for isotopes of chromium, platinum, and osmium are given. Isotopes of chromium are separated from perchloric acid solution as the trioxide, reduced with hydroxylamine sulfate, precipitated as the hydroxide, and ignited to Cr_2O_3. Platinum isotopes are precipitated as ammonium chloroplatinate, which is ignited, and the resultant platinum sponge is further ignited in hydrogen. Isotopes of osmium are volatilized as OsO_4, collected in hydrobromic acid, precipitated as hydrated OsO_2, and reduced with hydrogen to metallic osmium, the preferred inventory form.

This is the second article of a series on the chemistry of enriched electromagnetically separated stable isotopes enriched at Oak Ridge National Laboratory (ORNL). In the preceding issue of *Isotopes and Radiation Technology*,[1] the general problems and methods associated with the handling and purification of isotopically enriched materials and with their chemical and isotopic analyses were described. The purification methods for isotopes of strontium, barium, lead, zirconium, and hafnium were also presented. In this issue the purification methods for isotopes of chromium, platinum, and osmium are given.

CHROMIUM

Chromium has four naturally occurring stable isotopes with abundances of:

Isotope	Abundance, at.%
^{50}Cr	4.31
^{52}Cr	83.76
^{53}Cr	9.55
^{54}Cr	2.38

In the chemistry associated with the electromagnetic separation of chromium isotopes, described by Weaver et al.,[2] chromium is separated from perchloric acid solution as the trioxide, reduced with hydroxylamine sulfate, precipitated as the hydroxide, and ignited to Cr_2O_3, the stable inventory form (Fig. I-1).

Graphite collectors are now used exclusively in the enrichment of chromium isotopes. Although both copper and graphite pockets have previously been used to collect the isotopes of chromium, the difficulty of dissolving the chromium deposit from the copper pockets, and subsequently removing the relatively large quantity of copper that dissolves, has precluded the further use of copper.

The graphite collectors are cleaned on the outside by scraping with a half watch glass or with emery cloth and are then crushed and ignited at 600 to 800°C in either air or oxygen. The chromium oxide contains some residue from the graphite and other impurities, e.g., copper and stainless steel, from the calutron.

The residue from the graphite ignition is boiled in 70% perchloric acid, and the chromium, now in the hexavalent state, crystallizes out as

CrO_3 when the solution cools. The crystals are filtered on sintered glass, washed with 70% $HClO_4$, and dried with suction. Metals that have soluble perchlorates are removed by this treatment.

The suction flask is emptied, cleaned, and reconnected to the filter funnel containing the CrO_3. The residue is dissolved in distilled water, and the solution, which includes the filter washings, is collected in the suction flask. Any silica impurity remains on the filter. Any aluminum that is present in the product is precipitated with ammonium hydroxide and removed by filtration.

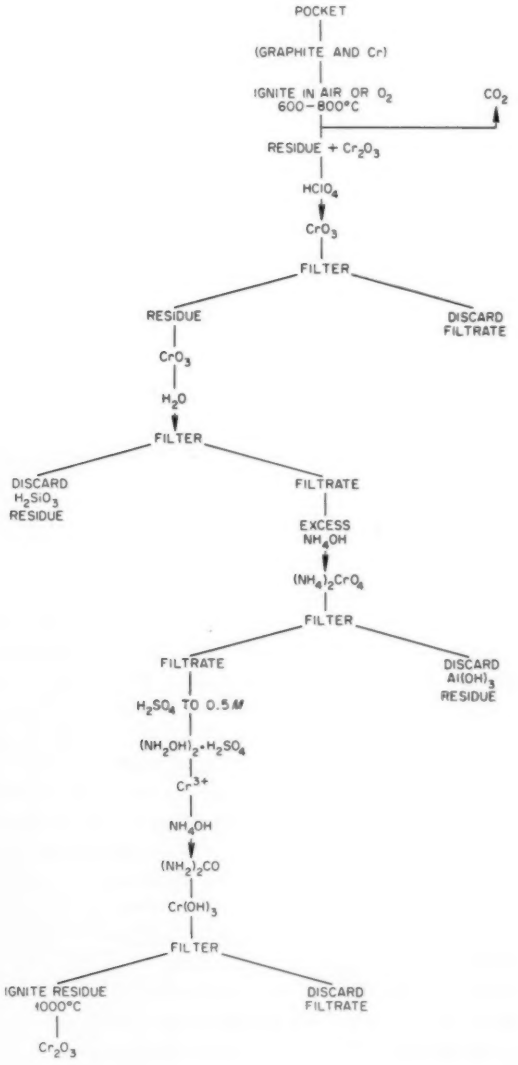

Fig. I-1 Purification method for enriched isotopes of chromium.

The ammonium chromate filtrate is neutralized, then acidified to approximately 0.5*M* H_2SO_4, and finally the chromium is reduced to the tripositive state by addition of hydroxylamine sulfate. Chromium is precipitated as $Cr(OH)_3$ by carefully raising the *p*H of the warm solution to near neutrality with 1 : 1 filtered ammonium hydroxide and then adding sufficient urea to precipitate the chromium. The precipitated $Cr(OH)_3$ is filtered on a fast paper, washed with hot water containing a small amount of NH_4NO_3, and ignited in porcelain or Vycor at 1000°C to Cr_2O_3, the inventory form. Only a few milligrams of this costly product, as nearly representative of the batch as possible, is used for chemical and isotopic analysis.

PLATINUM

Platinum has six naturally occurring isotopes, five of which are stable; one, ^{190}Pt, is an alpha emitter with a half-life of 6×10^{11} years. The natural abundances of the platinum isotopes are:

Isotope	Abundance, at.%
^{190}Pt	0.0127
^{192}Pt	0.78
^{194}Pt	32.9
^{195}Pt	33.8
^{196}Pt	25.3
^{198}Pt	7.21

In the chemistry for the purification of platinum isotopes, described by Davis and Sheridan,[3] platinum is precipitated as ammonium chloroplatinate, the precipitate is ignited, and the resultant "sponge" is further ignited in hydrogen (Fig. I-2).

Graphite pockets are used for collection of platinum isotopes and are thoroughly cleaned on the outside prior to crushing between cellulose acetate sheets and igniting in a quartz boat at 650°C in oxygen. The residue containing the platinum is dissolved in aqua regia and evaporated to incipient crystallization with excess hydrochloric acid. The resulting salts are alternately moistened with 12*M* HCl and dried to destroy any residual nitrate. The residue is then dissolved in 1*M* HCl with gentle warming, and the solution is filtered to remove any siliceous residue. After the solution has cooled,

solid ammonium chloride and then a volume of 95% ethanol equal to that of the solution are added to precipitate the platinum as the yellow ammonium chloroplatinate. The precipitate is allowed to settle for 24 hr and then is filtered, washed with 20% ammonium chloride solution, dried, and ignited at 800°C. The resulting platinum sponge is placed in a quartz tube and ignited to constant weight at 600°C in hydrogen. This is the stable inventory form.

OSMIUM

Osmium has seven natural isotopes with abundances as shown in the tabulation at the top of the next column.

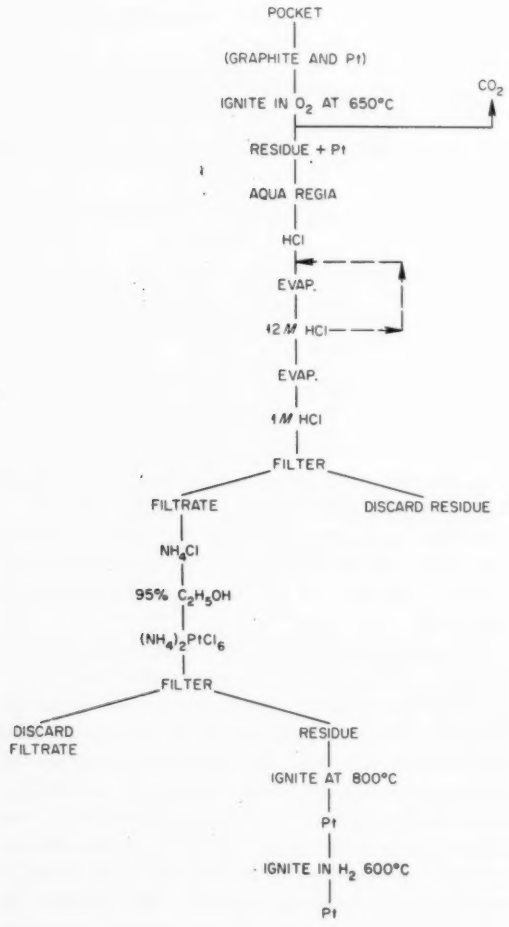

Fig. I-2 Purification method for enriched isotopes of platinum.

Isotope	Abundance, at.%
^{184}Os	0.018
^{186}Os	1.59
^{187}Os	1.64
^{188}Os	13.3
^{189}Os	16.1
^{190}Os	26.4
^{192}Os	41.0

In the chemical refinement of osmium isotopes described by Johnson et al.,[4] the graphite pocket with its deposit is ignited in oxygen, and the volatilized osmium is collected as OsO_4 in hydrobromic acid solution, precipitated as hydrated OsO_2 with sodium bicarbonate, and reduced to the metal with hydrogen (Fig. I-3). Although metallic osmium is not toxic, its volatile oxide, OsO_4, is known to create temporary blindness and to induce other undesirable effects;[5] thus it is necessary to take precautions when working with osmium.

Osmium isotopes are collected in graphite pockets, which are cleaned by buffing with abrasive cloth and then crushed in a plastic sheet. The graphite is ignited at 700°C in oxygen in a quartz boat and quartz furnace, which has a train of four traps connected to its exhaust. The first two traps contain 48% hydrobromic acid; the third contains 24% hydrobromic acid; and the fourth, inserted as a precautionary measure to reduce any OsO_4 that might pass through the other traps, contains a dilute solution of sodium hydroxide and a small amount of ethanol. The ignition temperature of the furnace is gradually raised to 700°C, and volatilized osmium is collected, essentially all of it in the first two traps. After overnight ignition the furnace is cooled to about 300°C and flushed with nitrogen. The first trap is then replaced, and the ignition in oxygen, at 800°C, is continued until the graphite is completely ashed. Any remaining OsO_4 is swept into the traps with nitrogen as before.

The hydrobromic acid solutions containing the osmium are combined and evaporated to near dryness on a steam bath. The residue is dissolved in a minimum of hot concentrated hydrochloric acid, and the solution is diluted and filtered. To the heated filtrate containing the osmium, sodium bicarbonate is added to approximately *p*H 6 to precipitate hydrated OsO_2. The oxide is centrifuged and washed, first with dilute ammonium chloride solution until no trace of sodium remains and then with concentrated

ammonium chloride solution to prevent ignition of the hydrogen used for subsequent reduction of the oxide to the metal.

The OsO_2 precipitate is heated at a low temperature until nearly dry, sampled for preliminary mass analysis, and then transferred to a quartz boat. The boat is placed in a quartz ignition system, which is flushed with carbon dioxide before hydrogen is admitted to reduce the OsO_2 to the metal at 900°C. The system is again flushed with carbon dioxide during cooling. Removal of adsorbed hydrogen by flushing with an inert gas such as carbon dioxide during the cooling process is considered to be important in stabilizing osmium metal for storage.[6,7] Osmium metal is the inventory form.

(R. E. Greene)

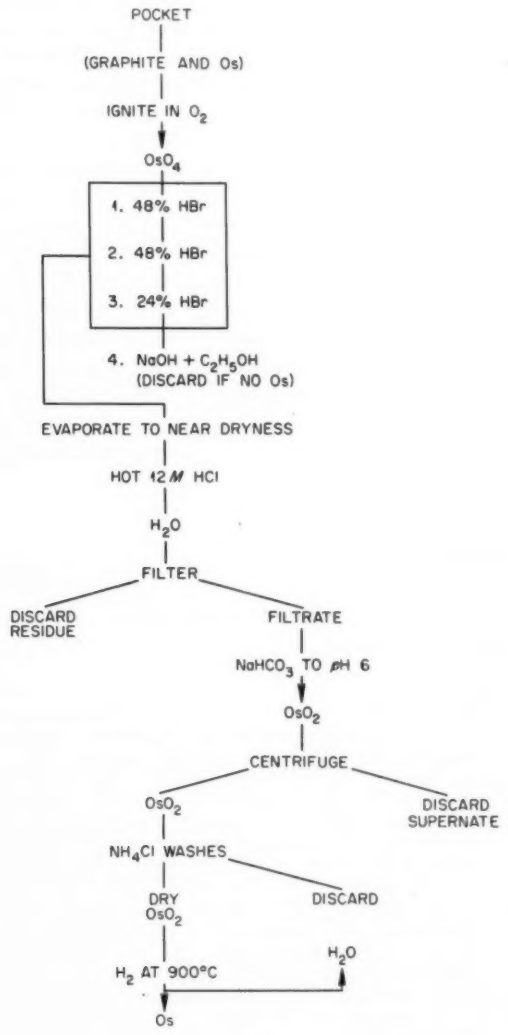

Fig. I-3 Purification method for enriched isotopes of osmium.

Radioisotope Production by Nuclear Explosion*

Analysis of the results of the Project Gnome isotope program indicated that the economics of obtaining the transplutonium elements, in particular berkelium and californium, by nuclear detonation will depend on the need for them and on the cost and availability from the ORNL reactor program. If still heavier elements can be produced in this way, their cost must be weighed against their value in research since there is no reasonable alternative method of production. Plutonium and lighter elements would be much more expensive than when obtained from reactor-irradiated material.

The purposes of the isotope program of Project Gnome, in which a nuclear explosion was carried out in a cavity in a salt deposit near Carlsbad, N. Mex., were to study the distribution of ^{242}Cm, ^{241}Am, ^{239}Pu, ^{228}Th, ^{210}Po, ^{204}Tl, and ^{3}H in the salt and the means by which these nuclides might be recovered. The finding of species with atomic mass 256 in the debris of an earlier test had suggested that transplutonium elements might be produced in weighable amounts by the use of nuclear explosives. However, their concentrations in the "ore" would probably be 0.1 ppm or less, and, unless they should happen to be concentrated in an easily accessible portion of the magma, their recovery would be expensive. Any tritium formed would be diluted by water present, and any reaction of tritium with oxidizing components in the melt or exchange with protium of water would aggravate the recovery problems. The Gnome project offered an opportunity to investigate these problems. Since it was not a production experiment, the isotopes studied were either a part of the nuclear explosive or were added in large enough amounts for subsequent determination.

*Summary of Isotope Program: Final Report, Project Gnome, M. W. Nathans, USAEC Report PNE-102F, Lawrence Radiation Laboratory, January 1965, 102 pages.

Data from analysis of the rock samples after detonation indicated that the most important minerals formed by reactions of the rock with the atmosphere and with materials introduced into the shot room or by reactions among the rock components were magnetite, olivine with and without iron, kirschtenite (or monticellite) with considerable iron substitution, and lead compounds. Alpha activity was associated primarily with the magnetite, olivine, and kirschtenite. Lead compounds carried no activity, quartz carried little or none, and white olivine carried little. The activity of the olivine appeared to increase with increasing depth of color.

Activity was distributed in the lower third of the cavity but inhomogeneously; i.e., the specific activities of melt samples varied by more than two orders of magnitude. Water leaching decreased this variation by an order of magnitude. Considerable amounts of strontium and cesium activity but less than 1% of the alpha activity appeared in the leach liquor.

Plutonium seemed to be embedded in an olivine matrix. Correlation between the iron and the plutonium (and curium) contents was good, although such was not the case for samples of material taken outside the cavity (tunnel and clay seams). Hydrochloric and nitric acids were equally effective in leaching the residues from the aqueous leach, but sulfuric acid was much less effective. Acid leaching rates appeared to be determined by the rate of hydrolysis of the olivine type compounds; after 1 hr, in different samples, 20 to 90% of the plutonium was solubilized, and probably essentially all could be in a sufficiently long time. The large amounts of silica gel that formed in some samples retained 10 to 15% of the leach solution.

Thorium, uranium, and curium behaved more or less like plutonium.

Less than 1% of the tritium present was found as HT, the rest being as HTO. The noncondensable gases were diluted by gases originating from the medium itself, and rubble from the partial collapse of the cavity contributed greatly to the amount of steam released. Tritium production by nuclear explosion would be economical only if the cavity atmosphere could be completely recovered, if the medium contained only a minimum amount of water, and if the cavity temperature remained high enough to keep the water in the gas phase. A major cavity collapse would probably preclude successful tritium recovery.

Fractionation of fission products, particularly gaseous elements and those with gaseous precursors, was considerable. This was also the case with the relatively volatile heavy elements thallium and polonium. Production of such elements by nuclear explosion would be uneconomical because they would be dispersed in too large an amount of melt and rubble, with a concentration of less than 10 ppb.

Some of the fractionation data indicate the following approximate time sequence of events: The most refractory components started to condense within a few tenths of a second, causing depletion of the cavity atmosphere in iron, the ^{144}Ce chain, actinides, and some other elements. A large fraction of the condensate was incorporated in debris and rock, which was never gaseous but which passed through the cavity in turbulent motion. Much of this material had time to collect near the bottom of the cavity. Shortly after condensation started, probably within 0.5 sec, the cavity contents found a path to the tunnel. Before the smog in the cavity could settle, the walls started to implode and to collect the smoke particles. Melt still adhering to the walls was entrained, and the falling debris caused melt to splash up and become incorporated in the bulk of the debris. Finally, the roof collapsed. (Martha Gerrard)

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Section

II

Isotopes and Radiation Technology

Isotope Technology Development

The review of the applications of Kryptonates, which was started in the preceding issue, is completed here with a discussion of the applications of Kryptonates in materials research. The isotopic method for determining enzyme activity is reviewed in another paper. The successful use of this technique in quantitative biochemical studies of microgram and submicrogram quantities of tissues suggests that, with further work to increase the sensitivity, the technique may be applicable to studies at the single-cell level. The use of gamma activation for analyzing rocks and ores is discussed in a third paper. Analysis by gamma activation is fast and permits determination of many elements that are not satisfactorily determined by conventional neutron activation analysis.

Applications of the Kryptonates in Materials Research

In the first of two articles,[3] the uses of Kryptonates in chemical analyses, as well as the preparation and properties of the Kryptonates, were reviewed. In the following review of the applications of Kryptonates in materials research, both feasibility studies and in-field applications are included. The uses of Kryptonates in the study of some factors related to friction, including surface temperature, oxidation, and wear; high-temperature oxidation kinetics; corrosion; photodegradation; phase changes; and solids under stress are discussed.

It has been predicted that it is probably in the area of materials research that the Kryptonates will provide the greatest advance in isotopes technology.[1,2] The preparation and properties of the "Kryptonates," as well as their uses in chemical analyses, were reviewed in the preceding issue of *Isotopes and Radiation Technology*.[3]

Kryptonates are solids — elements, inorganic and organic compounds, and alloys — that have been impregnated with a mixture of approximately 5% ^{85}Kr ($T_{1/2}$ 10.7 years, 0.695-Mev β) and approximately 95% stable isotopes of krypton either by ion bombardment or by diffusion under high temperature and pressure. The Kryptonates remain stable at room temperature, barring surface reactions; they are remarkably stable at elevated temperatures; and they are sensitive to chemical or physical disturbances at the surface of the solid, with these disturbances leading to proportional losses of ^{85}Kr.

Chleck, Cucchiara, and coworkers at Parametrics, Inc., introduced the Kryptonates and, under the auspices of the Atomic Energy Commission, Division of Isotopes Development, and the United States Air Force, Wright-Patterson Air Force Base, have done most of the work on the use of Kryptonates both in materials research and in chemical analyses.

In the following discussion the friction studies are in-field applications, and the remaining studies demonstrate feasibility applications of the Kryptonates in materials research.

Friction

Friction may be viewed with ambivalence. "If the coefficient of friction were less than it is, walking would be impossible and we should have to use cog wheels or perhaps some system of suction pads like an octopus to get about at all. Conversely, if it were much higher than it is, we should stick fast wherever we were put."[4]

The energy that is lost through friction is dissipated mainly in the form of heat and results in an increase in temperature. The measurement of this temperature is an acute prob-

lem in the design and operation of engine components. Such considerations as the construction materials, configuration, and lubrication and cooling systems of an engine component can be properly evaluated only if the temperature of the component is accurately known. Other measurement techniques demand that the components be accessible. The advent of the Kryptonates has provided a previously unobtainable measure of temperature, knowledge of which is basic to the understanding of the mechanism of friction and the associated mechanism of wear.

The development of this unique method for determining surface temperature is made possible by the fact that, when a Kryptonate is heated to a specific temperature, it loses a predictable fraction of its initial activity and there is no further loss of activity with further heating at or below this temperature. This means that the previous maximum temperature attained by a kryptonated object can later be determined by reheating and monitoring for radioactivity release. The onset of ^{85}Kr release marks the passing of the initial maximum temperature. Chleck, Maehl, and Cucchiara[5] have demonstrated the feasibility of several variations of the Kryptonate method for determining maximum temperature. These variations attempt to evaluate separately surface temperature and other factors of friction that chemically or physically disturb the surface of the solid, e.g., oxidation and wear, and contribute to the loss of ^{85}Kr.

MEASUREMENT OF SURFACE TEMPERATURE

Gas turbine engines are being operated with gas temperatures higher than the melting point of the turbine-blade material; only internal air cooling of the hollow blades prevents fusion. And military users of such power plants are demanding even greater power outputs. Thermodynamically this requires still higher operating gas temperatures.

By expanding their studies[3,5] on the thermal stability of Kryptonates at elevated temperatures, Chleck and coworkers[6] sought to determine the accuracy and precision with which temperatures above 1500°F could be measured by the kryptonation technique. High-temperature materials, such as boron nitride, type 304 stainless steel, tantalum, and titanium, were used. To determine the accuracy of the method under corrosive conditions, oxidation of the samples was permitted.

Kryptonated samples were heated in a furnace at a known high temperature for 10 to 20 min, which is sufficient time for equilibrium to be established. The samples were cooled and then reheated in stages (20 min at each temperature) in the same furnace. During both the preheating and the reheating, the furnace temperature was determined by Chromel–Alumel thermocouples read out on a Sim-ply-trol pyrometer with 50°F subdivisions from 0 to 2500°F and with a stated accuracy of ±2%. Thus, in evaluating the kryptonation technique, errors up to ±2% may be attributed to the thermocouple–pyrometer system. After each successive temperature stage, the samples were removed from the furnace and the radioactivity of each side of the sample was counted under fixed geometry with an end-window Geiger tube and scaler readout.

The results showed the sample activity to be constant until the previous maximum temperature was exceeded and further ^{85}Kr was lost. Additional activity loss was noted at each successively higher temperature. A plot of activity vs. temperature (part *a* of Fig. II-1) permits determination of the previous maximum temperature. The intersection of the steeply sloping line showing additional activity loss with the horizontal line showing constant activity (i.e., no loss) determines the previous maximum temperature. This basic pattern of loss on reheating has been observed for all the Kryptonates tested thus far.[6]

The plot in part *a* of Fig. II-1 of temperature measurements of titanium represents the results for the two sides of one sample. Two sets, each comprising three samples of kryptonated titanium foil, were preheated in an air environment to 1650 and 1725°F, respectively, for 20 min and then subjected to the reheating, also in an air environment, and counting procedures. The previous temperatures as determined by the intersections of the lines for the two sides of each of the three samples of the 1650°F preheat set were 1654 and 1659, 1655 and 1648, and 1652 and 1654°F. The mean value is 1654°F, and the standard deviation is only 3.6°F. The calculated accuracy (relative error = 0.24%) and the calculated precision [relative standard deviation (R.S.D.) = 3.6/1654 = 0.22%] point to an excellent temperature-measurement technique, which is limited by the

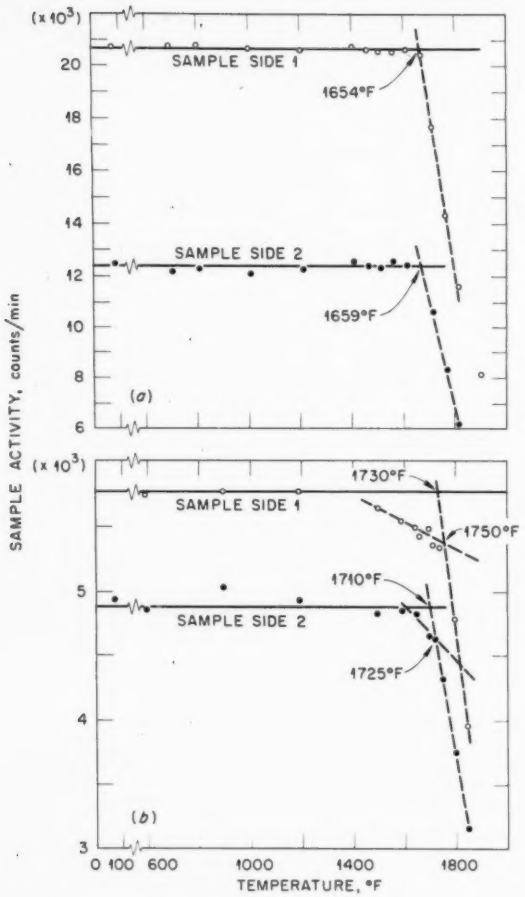


Fig. II-1 Temperature measurements of titanium samples preheated to (a) 1650°F and (b) 1725°F.

auxiliary temperature-measurement device rather than by inherent inaccuracy or non-reproducibility.[6]

In the determination of previous maximum temperature of the two sides of each of the three samples of the 1725°F preheat set, there were two intersections on the plots for each of the six cases. The results for the two sides of one sample are presented in part *b* of Fig. II-1. In addition to the intersections (in the vicinity of the previous maximum temperature) of the lines showing constant activity with the steep lines showing activity loss, there is, in each case, a second break in slope in the 1500 and 1600°F region. These lines of shallow slope continue until the steep activity-loss curves begin. Chleck[6] suggests that this trend, which appeared in all six cases in the 1725°F preheat set, represents continued oxidation* and that the following two choices for the temperature determination are available:

1. The slightly negative slope in the 1600 to 1725°F region may be ignored and the previous maximum temperature determined by the intersection of the best horizontal line with the steep thermal loss curve. The mean value is 1719°F, and the standard deviation is 10°F. The accuracy (relative error = 0.35%) and the precision (R.S.D. = 0.58%) are excellent.

2. A straight line may be drawn through the points of the shallow slope, and the intersection of the two *sloping* segments may be used. The mean value is 1736°F, and the standard deviation is 14°F. The accuracy (relative error = 0.64%) and the precision (R.S.D. = 0.81%) still fall within the limits of accuracy of the temperature-measurement device.

Although the kryptonation technique is capable of determining maximum temperatures even if oxidation occurs during the reheating cycle, as shown in part *b* of Fig. II-1 in which the loss in activity due to further oxidation of a kryptonated part is considerably less than the amount of activity lost at temperatures exceeding the maximum exposure temperatures, the technique is also capable of determining the previous maximum temperature minus any activity loss due to oxidation for samples such as those in the 1725°F preheat.[7] If the kryptonated part is reheated in an inert atmosphere, wear and/or oxidation do not occur, and the surface temperature is the only cause of activity loss.

The established reproducible behavior[3] of Kryptonates prepared either by ion bombardment or by diffusion is further verified by the activity loss with temperature and at reduced pressures of three bombardment-kryptonated tantalum wires (Fig. II-2). This reproducible behavior permits the use of the data as a calibration curve for temperature measurements in the absence of reaction. Thus, bombardment-kryptonated tantalum-wire samples may be counted during or after the initial heating and the temperature read from the calibration curve. An analysis of the data showed that this method may be capable of

*In a personal communication, May 17, 1966, Cucchiara suggests that, in view of the fact that severe oxide flaking was observed at temperatures greater than 1800°F, the additional 75°F increase was sufficient to cause minor flaking and/or lifting of the protective oxide film. On reheating, further oxidation occurs with a resultant second slope.

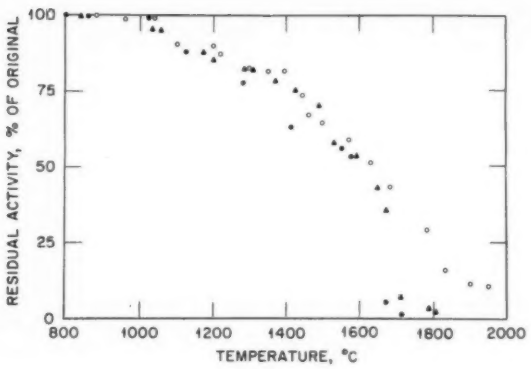


Fig. II-2 Reproducibility of activity loss of three bombardment-kryptonated tantalum wires.

measuring temperature to ±30°C or better (to at least 2000°C). The applicability of this method is not limited to tantalum wires, although specific calibrated outgassing curves would be necessary for each type of material.

Since the kryptonated boron nitride samples showed a premature loss similar to that of the titanium 1725°F preheat set and since the kryptonated stainless-steel (type 304) samples underwent severe destructive oxidation at approximately 1500°F, both materials were abandoned.

Chleck and coworkers[6] further sought to demonstrate that the kryptonation technique permits the determination of the maximum temperature attained by portions of the surface of a component that has experienced a variation in maximum temperature over the surface. Thermocouples were attached to a kryptonated 6-in.-long by ¼-in.-diameter aluminum rod at five equidistant points (1 in. apart and 1 in. from each end). With a 2½-in. length inside the furnace and a 3½-in. length outside the furnace and cooled by a blower and heat sink of copper in contact with liquid nitrogen, the rod was preheated to produce a surface temperature gradient and held for 50 min to ensure equilibrium. The reheating and counting procedures were applied to the five portions of the rod to determine the distribution, or profile, of previous maximum temperatures over the surface. The counting was limited, by lead shielding, to ½-in. lengths centered about the thermocouple locations.

At each numbered thermocouple the previous maximum temperature (Table II-1) was determined by the intersection of the horizontal line of constant activity with the sloping line of activity loss. The maximum error of less than 2.5% is satisfactory in view of the ±2% uncertainty of the thermocouple–pyrometer system and in view of the fact that each area counted actually experienced a temperature gradient *within* its boundaries. The largest errors are associated with thermocouples Nos. 4 and 5, which are in areas with greatest temperature gradient.

The ultimate test of the value of the above technique was the determination, by Chleck and coworkers[6] in conjunction with an engine manufacturer, of the maximum temperature profile over the surfaces of turbine blades mounted in an operating engine.

Table II-1 PREHEAT AND PREVIOUS MAXIMUM TEMPERATURES OF PORTIONS OF AN ALUMINUM ROD

Thermocouple No.	Temperature, °C		
	Preheat	Gradient	Previous maximum determined by kryptonation technique
5	550		560
		95	
4	455		465
		70	
3	385		380
		60	
2	325		325
		25	
1	300		305

Four 2½- by 1-in. turbine blades were kryptonated at 1000 psi and 400°C for 10 hr, removed from the apparatus, and counted to ensure achievement of a sufficient activity level. The counting apparatus consisted of an end-window Geiger tube, a lead shield with a ⅜-in. slit, and a movable mount for the blade. The Geiger tube, the slit in the lead shield, and the section of the blade being counted were aligned along a single axis, thus permitting the counting of only the area of interest (a ⅜-in. section). The blades were then mounted in a jet engine, which was operated for about 3 hr. Under these conditions activity was lost from the blades in an amount dependent upon the maximum operating temperature of the engine (i.e., the preheat temperature). The blades were then removed from the engine for temperature determination.

Under controlled conditions each blade was reheated in 100°F increments up to temperatures 300°F higher than the maximum prestress temperature (the maximum temperature to which the component had been subjected in the initial heating cycle, i.e., in the engine).

The blades were held at each temperature for 20 min, removed from the oven, cooled, and the residual activity was counted. The temperatures at seven selected areas of the convex side of each blade were determined. The maximum temperature was determined from a graph of counts per minute vs. temperature using the intercept of the horizontal and sloping lines. The method of least squares was used to fit the straight lines.

Figure II-3 shows a temperature profile obtained on an actual blade shape, and Fig. II-4 shows temperature profiles obtained for two of the blades that were identical in composition and hence were expected to demonstrate the same heat-transfer characteristics. Temperature data are given as a fraction of the maximum temperature value obtained for the given blade.

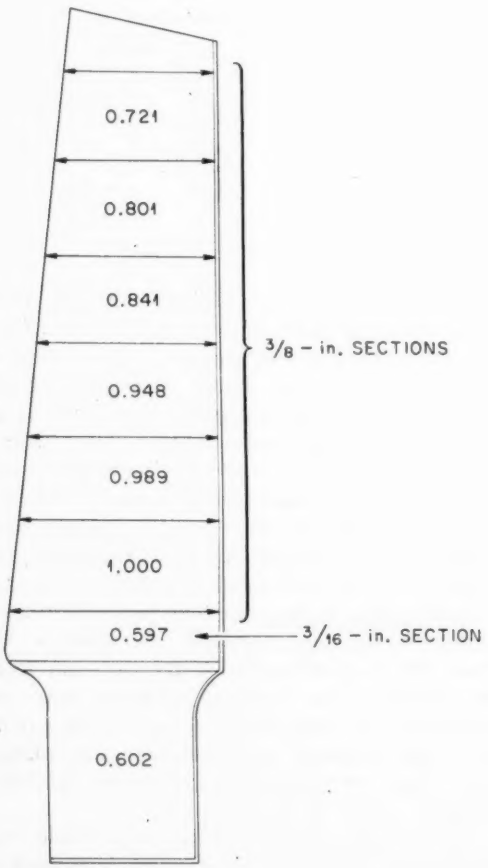


Fig. II-3 Temperature profile on actual turbine blade shape. The data are presented as a fraction of the maximum temperature value obtained for the blade, T/T_{max}.

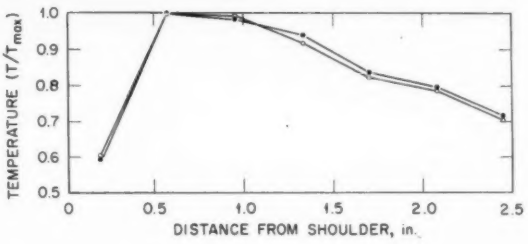


Fig. II-4 Temperature profile for two turbine blades of identical composition and for the turbine blade in Fig. II-3.

Cucchiara and Goodman[7] suggest that the two factors that determine the accuracy of the technique are the magnitude of the temperature interval in the reheating cycle and the counting statistics. The accuracy of the temperature determination increases as the size of the temperature interval decreases. The accuracy has been shown by a large number of temperature determinations to be ±25°F for 100°F temperature intervals and ±10°F for 50°F temperature intervals. Unless counting times are very lengthy, samples of low initial activity yield statistical deviations so great that the temperature breakpoint is completely masked. Also, the higher the initial activity of the sample, the smaller is the surface area over which the temperature can be determined. For high-activity samples (>1 μc/sq in.), previous maximum temperatures have been determined for sample areas as small as 0.03 sq in.

Wisnieff and Bardach[8] propose that, for determining maximum temperatures, a gas-evolution method of counting the activity is superior to the residual-activity method in that more extensive counting data can be accumulated in a shorter period of time. However, if temperature measurements on various areas of a kryptonated material are sought, as for a temperature profile of a turbine blade, it is necessary that the material be cut into small sections and thus destroyed.

OXIDATION

The formation of surface oxides or other corrosion products on two solid surfaces that are sliding over each other considerably influences the friction process. The Kryptonates are sensitive to chemical or physical dis-

turbances of the surface of the solid, and such disturbances lead to proportional losses of ^{85}Kr. Thus, if an oxide or corrosive film is formed through friction, the rate of formation of this film can be determined by the loss in ^{85}Kr.

The rate of oxidation and the surface temperature will have a combined effect on the release of ^{85}Kr from a kryptonated source. The capability of the kryptonation technique to determine maximum exposure temperatures even if oxidation occurs during the reheating cycle was illustrated in part *b* of Fig. II-1. To determine ^{85}Kr loss through oxidation alone,[7] the kryptonated sample is thermally stabilized by heating in an inert atmosphere to a temperature approximately 100°F higher than the average maximum surface temperature expected during the oxidation process. No significant loss in activity due to temperature will be observed during reheating at or below the stabilizing temperature. In the absence of wear and of surface temperature in excess of the stabilizing temperature, the loss of sample activity is due entirely to oxidation and is directly proportional to the rate of the reaction.

Cucchiara and Goodman[7] report the experimental verification[9] of the applicability of Kryptonates to delineate, by either of two techniques, the oxidation or corrosion processes in materials. The techniques parallel the gas–solid reactions of the hazardous-vapor determinations reviewed[3] in *Isotopes and Radiation Technology*. In one technique the central element is a kryptonated solid that will react with the test gas of interest (part *a* of Fig. II-5) and

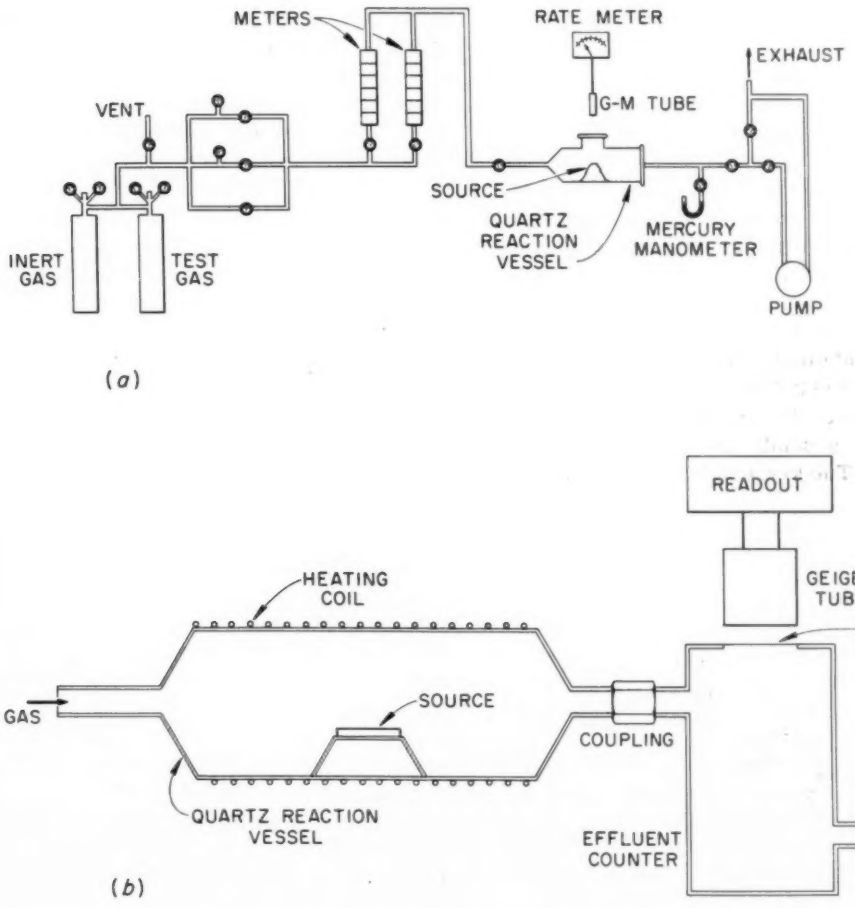


Fig. II-5 Apparatus for two gas-solid reaction counting techniques. (a) Source is counted for residual activity. (b) The effluent activity is counted.

rate of loss of activity from the source is a measure of the rate of the reaction. In the second technique the effluent activity, i.e., the gaseous activity removed from the source, is counted (part *b* of Fig. II-5). Effluent-activity counting is particularly advantageous when the source cannot be counted directly. By either technique, the ^{85}Kr loss is directly proportional to the rate of the oxidation reaction.

WEAR

The removal or wear of the surface of a kryptonated source will result in a decrease in the residual activity of the sample. To obtain ^{85}Kr loss from wear alone, the kryptonated sample is preheated to a temperature higher than the expected average maximum temperature during the wear process and, after wear, is reheated in an inert atmosphere. The loss in activity will be solely a function of the amount of wear.

Preliminary studies by Cucchiara and Goodman[7] showed that the wear process can be followed in detail by application of the kryptonation technique. A kryptonated copper sample in the form of a 1- by 1- by $\frac{1}{2}$-in. stationary block is allowed to undergo sliding friction in the apparatus shown in Fig. II-6. The apparatus consists of the stationary kryptonated metal block in contact with a sliding metal block, which is actuated by a cam attached to a shaft on a motor. A stainless-steel rider is mounted on the moving block. The region of contact between the surfaces is clearly defined since it occurs between a small curved protrusion and a flat surface. The kryptonated copper block is continuously monitored to determine the loss in activity. The system is operated at 130 strokes/min with a head load of 250 g.

Activity loss is due to the combined effects of temperature, oxidation, and wear since in the preliminary experiments the kryptonated sample is not thermally stabilized and the wear process is allowed to occur under room ambient conditions. However, in the copper–steel system, wear is severe, and, for all practical purposes, the activity loss can be considered to be due almost entirely to wear. In Fig. II-7 the loss in activity follows a smooth curve with time, indicating that, for the particular system under study, wear follows a regular pattern.

Cucchiara and Goodman[7] also demonstrated that the technique can indicate wear irregularities or the sudden onset of severe wear such as a lubricant breakdown or sudden load increase. In this system a 1- by 1- by 0.005-in. kryptonated rough copper foil is mounted in the wear apparatus. A smoothly polished 0.1- by 2- by $\frac{1}{4}$-in. brass block replaces the stainless-steel rider of the above system. Since the foil is not smooth, the moving brass block does not make complete contact with the surface of the foil but rather rides on the projections of the foil until the projections are completely worn. The rate of activity loss is very mild until the projections are worn and complete contact of the two surfaces occurs. The rate of activity loss increases greatly once contact is made, indicating the onset of severe wear. This system is operated at 60 strokes/min with a head load of 875 g.

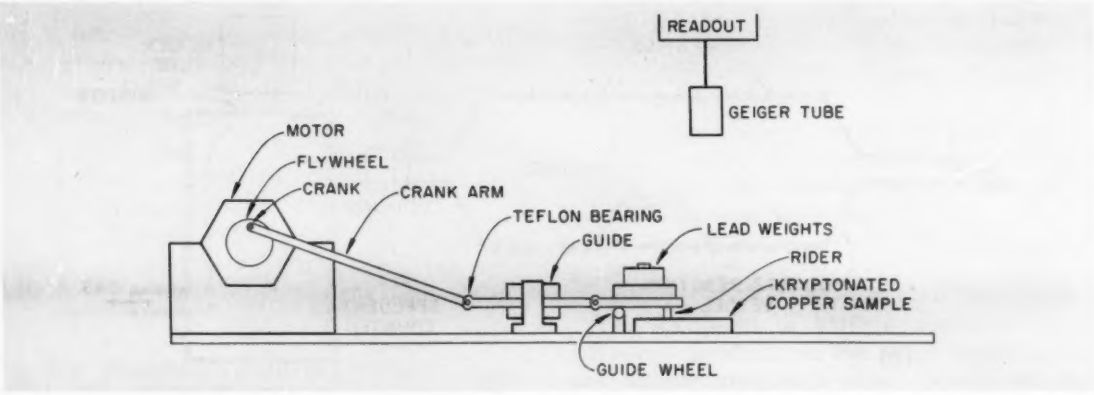


Fig. II-6 Apparatus for the subjection to, and measurement of, wear of a kryptonated sample.

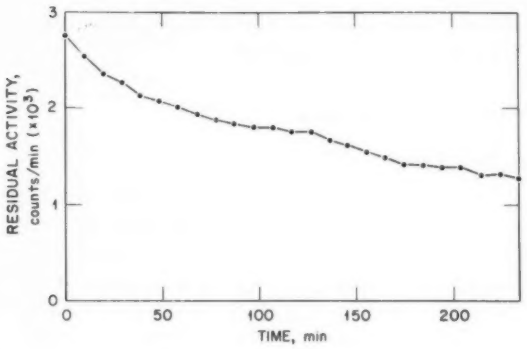


Fig. II-7 Activity loss for the kryptonated copper sample subjected to wear in the friction apparatus.

CONCLUSION

The determination of the previous maximum temperature by the kryptonation technique is currently being used in the collection of design data. Future plans call for use in the collection of inspection data. Numerous parts for production-run engines will be kryptonated, and during the periodic overhaul disassembly of the engines, the various kryptonated parts will be inspected to determine the operating conditions.

High-Temperature Oxidation Kinetics

Apart from the role of oxidation in the determination of surface-temperature measurements in friction studies, the high-temperature oxidation of refractory metals is a difficult and currently important area of investigation. Of particular concern are the changes in metals caused by conditions in outer space.

The sensitivity of the Kryptonates to chemical disturbances of the surface of the solid and the loss of ^{85}Kr in proportion to the disturbances are the properties basic to the high-temperature oxidation studies by Goodman and Ronayne.[10]

In the light-bulb type oxidation cell (Fig. II-8), a kryptonated refractory-metal wire sample, such as tantalum, is attached to two tungsten leads to permit resistance heating of the tantalum. The kryptonated sample is heated in the absence of oxygen to a temperature in excess of the oxidation temperature. When the activity loss at the predetermined temperature ceases, the wire is cooled to room temperature. Oxygen is then admitted through the adjustable leak valve, the sample activity is monitored through a thin aluminum-foil window mounted on the vessel, and continuous evacuation is accomplished by diffusion and/or mechanical pumps located on the downstream side of the wire. The wire is brought to reaction temperature without altering the measured constant oxygen leak rate or the constant mechanical pumping speed.

At the same time that the pressure gages, which are mounted on the downstream side of the wire, are monitoring the pressure during the course of the reaction, the optical pyrometer is measuring the wire brightness, i.e., temperature. If necessary in order to maintain constant brightness (temperature) during the course of the run, the voltage applied to the wire is varied.

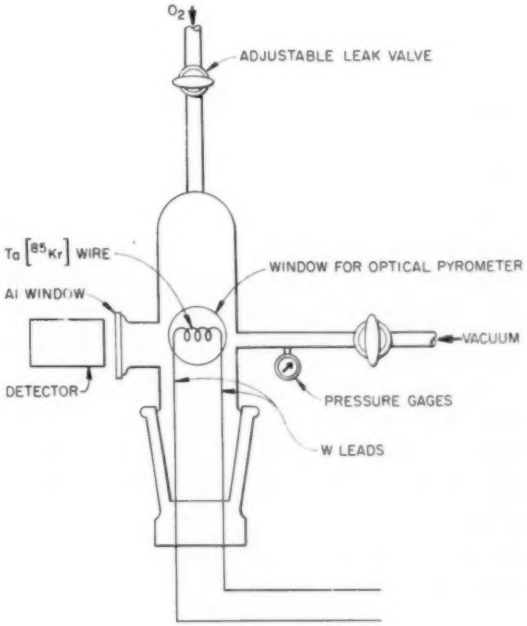


Fig. II-8 Light-bulb type oxidation cell.

Nine runs were performed at temperatures between 1400 and 1700°C. As evidenced by a run at 1600°C and an initial oxygen pressure of 10^{-3} torr (Fig. II-9), activity and pressure show parallel behavior. The increase in pressure at about 19 min followed by a pressure maximum at about 21 min corresponds to the decrease in the activity loss rate at about 16 min followed by an activity-loss rate increase at about 19 min. The pressure increase corresponds to a decreased rate of oxygen uptake. The pressure-change lag of about 3 min

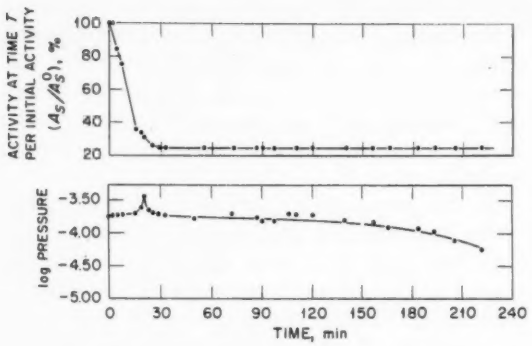


Fig. II-9 Activity and pressure as a function of time for a tantalum run at 1600°C and an initial oxygen pressure of 10^{-3} torr.

is attributed to a delay between the time the rate of oxygen uptake by the kryptonated sample changes and the time that enough oxygen has passed the wire to effect a change in pressure at the gages. The decreases in pressure observed by the pressure gages at zero time result from depletion of the oxygen leak, the leak rate being rather small, by reaction with the kryptonated tantalum sample; again, pressure is seen as a measure of oxygen uptake by the sample.

As expected, the initial activity loss rates generally increased with increasing temperature and pressure (Table II-2). On the basis of the results of the nine runs plus the results of two additional runs at 1800°C, Goodman and Ronayne conclude that the release of ^{85}Kr accurately follows oxygen uptake for $Ta + O_2$ reactions occurring at temperatures up to 1800°C and thus is an accurate reflection of tantalum oxidation kinetics.

Table II-2 INITIAL ACTIVITY-LOSS RATES FOR THE NINE SAMPLES OF THE NINE RUNS

Sample No.	Temp., °C	Initial oxygen pressure, torr	Initial $\frac{d(A_S/A_S^0)}{dt}$, %/min
1	1400	10^{-4}	0.46
2	1400	10^{-3}	1.41
3*	1400	10^{-2}	13.5
4	1600	10^{-4}	1.8
5†	1600	10^{-3}	4.0
6‡	1600	5×10^{-3}	32.0
7*	1600	10^{-2}	9.34
8	1700	10^{-4}	8.75
9*	1700	10^{-2}	8.25

*Runs made at 10^{-2} torr showed a not fully explained decrease in rate with increase in temperature.
†Activity losses and pressure changes with time plotted in Fig. II-9.
‡The observed initial activity-loss rate is exceptionally high and may not be meaningful.

Corrosion Studies

METALS IN SOLUTION

Some of the gas–kryptonated-solid reaction studies, such as the high-temperature oxidation kinetics of tantalum, may also be considered as corrosion-detection and corrosion-rate studies. To supplement these gas–solid corrosion studies, Chleck et al.[6] exposed kryptonated aluminum-alloy samples [Al 7075-T6 and Al 7075-T73 (tempered)] to 10% NaCl solutions.

The two sides of each alloy sample were counted, and the data were plotted as the activity loss vs. exposure time. The scatter among data points for the two sides of the same alloy sample was much greater for the tempered sample, as was expected because of the additional inhomogeneities introduced on the specimen surfaces by the tempering treatment. Whereas the activity loss for the Al 7075-T6 alloy continued for the duration of the test, the activity loss for the Al 7075-T73 alloy decreased markedly during the 20- to 100-min time interval. The conclusion that the Al 7075-T73 (tempered) alloy has a greater corrosion resistance was in accord with expectation based on prior experience.

PROTECTIVE COATINGS

The claim that a paint was protective to 425°C was verified by a study of the relative oxidation rates of painted and unpainted kryptonated iron samples. Whereas the unpainted sample was oxidized at a relatively rapid rate at the lower temperatures, the painted sample did not attain a comparable rate of oxidation until the temperature limit of protection was reached. The data not only show the type of results obtainable in the evaluation of a group of coatings for direct comparison purposes but suggest the value of Kryptonates in studies of the effect of environmental exposure on coating deterioration and on the breakdown of components, e.g., pigments, under test conditions.

Photodegradation Studies

Reflective coatings for aerospace vehicles are known to be degraded by photochemical reaction.[6] In a study initiated to establish the feasibility of using kryptonated homologs of

thermal control coatings to determine whether these homologs could be used to investigate degradation by ultraviolet irradiation, samples (e.g., titanium dioxide–silicone) were kryptonated and thermally stressed under simulated environmental conditions. The samples were then illuminated with ultraviolet radiation, and the activity losses were observed. The samples were shown to lose activity in a smooth fashion through ultraviolet irradiation, and the losses were ascribed to photochemical degradation. The kryptonation technique was shown to be more sensitive than the standard (measurement of spectral reflectivity) technique,[6] which is currently being used to investigate this degradation.

Detection of Phase Changes

Goodman and Ronayne[10] have demonstrated the feasibility of the kryptonation technique to detect crystalline phase changes. The rate of ^{85}Kr release from the sample indicates the occurrence of transition, as long as the transition involves a substantially large crystalline rearrangement.

SOLID–SOLID TRANSITION

On initial heating, a sample of CsCl, which is a body-centered cubic crystal that is known to undergo a transition to a face-centered cubic crystal at 445°C, started losing activity at about 445°C and continued to lose it, at a slower rate, as the temperature increased.

In an attempt to determine the feasibility of the technique for distinguishing activity loss due to phase change from that due to sample degassing, the CsCl sample was heated to 410°C and held there. The sample activity here decreased and then became essentially constant (Fig. II-10). When the sample was held at a constant temperature in the 445 to 465°C range — the interval of the suspected phase change — the activity decreased in a roughly exponential manner. When the activity level had become approximately constant, the temperature was increased and there was a further activity loss. But the activity loss ceased almost immediately when the heating was stopped at about 525°C. Whereas at constant temperature the activity loss was roughly exponential with time in the region of the phase change, it was constant at constant temperature during sample degassing.

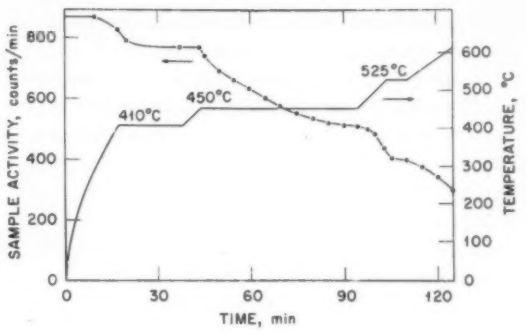


Fig. II-10 Phase change and degassing activity losses on heating kryptonated CsCl sample.

SOLID–LIQUID TRANSITION

Since the kryptonation technique is applicable only to transitions that involve a substantial crystalline rearrangement, solid–liquid transitions are more readily detected. The curve for Na_2CO_3 (Fig. II-11), which melts at 854°C, shows an abrupt change in slope at about 850°C. The Na_2CO_3 is reported to undergo three solid–solid transitions — at 356, 486, and 618°C — and none of the three reported transitions was detected.

Goodman and Ronayne[10] suggest that, with improvement of the apparatus, for example, by incorporation of a temperature programmer to provide a constant rate of temperature increase, further studies would delineate more closely the extent of the reconstructive transformation required for solid–solid transition detection and the accuracy to which the temperature of the transition may be specified.

Stress-Strain Studies

In preliminary experiments to demonstrate the value of the kryptonation technique in the study of solids under stress, Chleck et al.[6] subjected 0.006-in.-diameter kryptonated Nichrome wires in air at room temperature to tensile loads of 81,500, 110,000, and 120,000 psi. In the plots of source activity as a function of time, the time-dependent activity loss, which followed the application of tensile stress, paralleled expectations for the course of creep. In every case, almost all the activity was lost immediately, but the residual activity was higher for the wire subjected to the smallest stress. Thus the release of ^{85}Kr during the

resultant strain (deformation) at or near the surface was a measure of stress.

In another experiment, 0.006-in.-diameter kryptonated Nichrome wire in a nitrogen atmosphere at room temperature was subjected to a tensile load of 81,500 psi with no loss of activity. However, when the nitrogen was replaced by oxygen, the activity loss was immediate, which suggests that the loss in activity at room temperature in air is associated with oxidation of surfaces newly susceptible to oxidation as a result of the creep in the wire. Such information on the behavior of a solid actually under stress would be difficult to obtain by any other means.

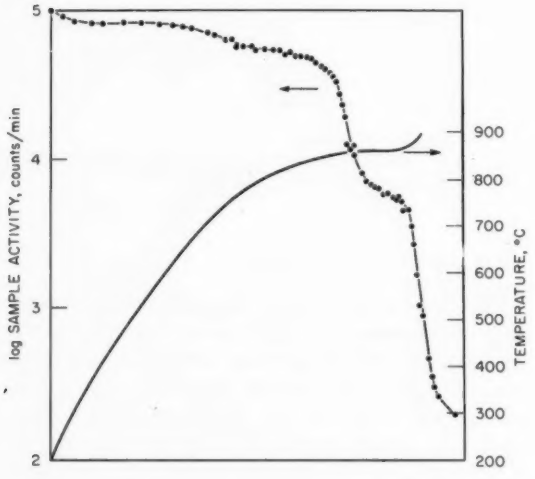


Fig. II-11 Phase change activity loss on heating kryptonated Na_2CO_3.

Conclusion

If only the studies reviewed were considered, the applications of the Kryptonates in materials research would be many; but plans call for the extension of the concepts. Not only that, but applications beyond these are anticipated as more is understood about properties such as the binding energy of the Kryptonates. It may well be that it is in the area of materials research that the Kryptonates will provide the greatest advance in isotopes technology.

(Joan E. Carden)

Isotopic Method for Determining Enzyme Activity in Microgram and Submicrogram Quantities of Tissue*

By Richard E. McCaman, Ph.D.†

The isotopic method for determining enzyme activity is reviewed, and representative protocols are presented. The enzyme preparation is incubated with a radioisotope-labeled substrate, and the radioactivity of the isolated reaction product is measured. The methods generally have a sensitivity of 10^{-12} mole of enzyme product when the substrates have a specific activity of 10 mc/mmole. It is suggested that these techniques may be applicable to metabolic studies at the single-cell level.

This review of the isotopic method for determining enzyme activity is a general consideration of the technique and its successful use in quantitative biochemical studies of microgram and submicrogram quantities of tissue. This method, in which the enzyme preparation is incubated with a radioisotope-labeled substrate and the amount of radioisotope-labeled product is measured by scintillation counting, is inherently sensitive and may be potentially useful in single-cell chemistry.

Metabolic studies at the level of a single cell‡ require highly sensitive, and frequently highly technical, methodology. Of the physical and chemical techniques theoretically capable of providing quantitative biochemical information at this level, some of them, e.g., X-ray diffraction and dark-field microscopy, have limited practical usefulness. The bulk of our current quantitative biochemical information at the level of single cells has been provided

*Adapted from a paper presented at the Symposium on Recent Developments in Research Methods and Instrumentation, Bethesda, Md., Oct. 4–7, 1965. Much of the author's personal research described in this presentation was supported by a grant from The National Multiple Sclerosis Society.

†Institute of Psychiatric Research, Indiana University School of Medicine, Indianapolis.

‡It should be emphasized that the author's research is devoted almost exclusively to metabolic studies of the nervous system. Although the general aspects of this presentation are applicable to metabolic studies with other tissues, some of the specific statements (weight of single cell, tissue limits, etc.) have specific relevance to nervous tissue.

by three techniques*: *ultraviolet microscopy*, which has been widely used in studying nucleic acid metabolism;[11] *gasometric* measurements, which have been used in studying the metabolism of substrates and the activities of discrete enzymes;[12] and *fluorescence* measurements, which have provided many interesting data on enzyme activities and levels of endogenous substrates related to pyridine nucleotide metabolism.[13] Of these three, the last appears to offer the broadest potential for quantitative study of the enzymes and substrates involved in metabolic pathways provided the system under study can be coupled (via the use of additional purified enzymes as auxiliary reagents) to one in which the nucleotide is oxidized or reduced. However, if the isotopic methods can be developed, they would have a sensitivity at least equal to that of the fluorescence procedure but would be directly applicable to the system under study. More important would be the use of isotopic procedures for the study of enzyme systems not accessible to study by the fluorescence (or any other) procedures. It seems likely that if the appropriate isotopic methodology could be developed to study the enzymes involved in the metabolism of only a portion of the labeled biologic compounds currently available from commercial sources, they would represent one of the most powerful tools for metabolic studies available to the biochemist.

Sensitivity and Cost of Isotopic Method

SENSITIVITY

The sensitivity of the isotopic method is generally 100 to 1000 times greater than that of colorimetric methods and is generally comparable to that of fluorimetric methods:

Isotopic	10^{-11} to 10^{-12} mole
Colorimetric	10^{-9} to 10^{-10} mole (micro)
Fluorimetric	10^{-10} to 10^{-11} mole (NADH, NAD)†

These values represent theoretical limiting sensitivities, and, for the isotopic method, the values are based on use of radioisotope-labeled substrates with specific activities of 1 to 10 mc/mmole. The sensitivity can be increased by using a radioisotope-labeled substrate with a higher specific activity.

The limiting sensitivity is calculated as follows: Assuming a substrate with a specific activity of 1 mc/mmole, we multiply the moles per millicurie by the ratio of millicuries to disintegration rate, which is a constant, and assume a counting efficiency of 70% (a conservative value) for ^{14}C:

$$\frac{10^{-3}\ \text{mole}}{\text{mc}} \times \frac{\text{mc}}{2.2 \times 10^{9}\ \text{dis/min}} \times \frac{\text{dis/min}}{0.7\ \text{count/min}} = \frac{6.5 \times 10^{-13}\ \text{mole}}{\text{counts/min}}$$

The value 6.5×10^{-13} mole/(count)(min) has the practical limitation that the equipment blank (noise level, emission from potassium in vials, etc.) is some 20 times larger. If we correct for this, we arrive at

$$\text{Limiting sensitivity} = 1.3 \times 10^{-11}\ \text{mole}$$

Therefore, at about 10^{-11} mole, the *net* count rate equals the equipment blank value. I use this mode of expression[14] so that the value may be compared directly with the values for NADH and NAD of 10^{-10} to 10^{-11} mole. A similar calculation[15] for a colorimetric method, assuming an extinction coefficient of 10,000 and a cuvette with a 1-cm light path (50 μl volume), yields a much lower sensitivity, i.e., 0.5×10^{-9} mole.

COST

Although the cost of radioisotope-labeled compounds is an important factor, particularly if a premium is put on purity, the isotopic method is not necessarily a costly one. One of our more expensive substrates, ^{14}C-acetyl CoA (see first protocol below), costs about $100 for 50 μc. However, because of the small amount used per analysis when working at the microgram level, several thousand analyses may be made with $100 worth of material.

Protocols for Enzyme-Activity Determination

When isotopic procedures are used for measuring enzyme activities, the biggest problem in achieving high sensitivity is separation of the

*For a comprehensive review of the various techniques (other than isotopic) referred to above and their application to various metabolic studies, see *Quantitative Histo- and Cytochemical Techniques*, D. Glick (Ed.), Vols. I and II, John Wiley & Sons, Inc., New York, 1961.

†The following abbreviations are used: NAD, nicotinamide dinucleotide; NADP, nicotinamide dinucleotide phosphate; NADH,NADPH, reduced forms of the respective nucleotides; ATP, adenosine triphosphate; CoA, coenzyme A; FDB, fluorodinitrobenzene.

labeled product of the reaction from the labeled substrate before the product radioactivity is measured. The amount of labeled product is usually 0.1 to 5% of the amount of labeled substrate, and the separation procedure used should not only allow quantitative recovery of the product but should also be conveniently and rapidly carried out. Various physical and chemical techniques, including chemical reactions of

Enzyme	Substrate	Derivative
Choline acetyl transferase	^{14}C-acetyl CoA (choline)	^{14}C-acetylcholine (reineckate)
Carnitine acetyl transferase	^{14}C-acetyl CoA (carnitine)	^{14}C-acetylcarnitine (periodide)
Choline phospho-kinase	^{14}C-choline (ATP)	^{14}C-choline phosphate (reineckate)
Ethanolamine phosphokinase	^{14}C-ethanolamine (ATP)	^{14}C-ethanolamine phosphate (FDB)
N-methyltrans-ferase	^{14}C-adenosylmethionine (norepinephrine)	^{14}C-epinephrine (reineckate)

Table II-3 TYPICAL ISOTOPIC PROCEDURES FOR ENZYME MEASUREMENT

Enzyme	Incubation (30 min at 38°C)	Postincubation conditions	Coefficient of variation, %	Limiting sensitivity,* mole
Choline acetyl transferase	0.1–20 μg of tissue† in 10 μl of buffer–substrate: K_2HPO_4 (7.4) 0.1*M* choline 5m*M* ^{14}C-acetyl CoA 0.05m*M*	Choline 0.2*M* 1 μl of trichloroacetate 3*N* CENTRIFUGE 10 μl into 20 μl of acetyl CoA 5m*M* in HCl 0.5*N* 7 μl of reinecke salt (saturated soln) CENTRIFUGE (discard supernatant) 50 μl of HCl (0.5*N*) to wash ppt. CENTRIFUGE (discard supernatant) 50 μl of acetone to dissolve ppt. COUNT ALIQUOT	3	10^{-12}
Monoamine oxidase	0.5–40 μg of tissue in 10 μl of buffer–substrate: K_2HPO_4 (7.2) 0.1*M* ^{14}C-serotonin 0.8m*M*	1 μl of HCl 3*N* 50 μl of ethyl acetate CENTRIFUGE 40 μl of organic layer 50 μl of HCl "wash" COUNT ALIQUOT	2–3	10^{-12}
Choline phospho-kinase	0.1–40 μg of tissue in 10 μl of buffer–substrate: aminopropanol (10.2) ATP 10m*M* ^{14}C-choline 20m*M*	20 μl of NaOH 0.4*N* 10 μl of reinecke salt (saturated soln) CENTRIFUGE 25 μl of supernatant 5 μl of choline 0.1*M* 10 μl of reinecke salt (saturated soln) CENTRIFUGE COUNT ALIQUOT	3	10^{-12}

*Based on assumption that the specific activity of labeled substrate was approximately 10 mc/mmole.
†Lesser quantities of tissue could be used if incubation time or specific activity of substrate were increased.

the product or substrate, solvent extraction, and microdiffusion, have been used to achieve these goals. A representative selection of our methods and those used elsewhere is presented in Table II-3 to illustrate the ease with which the level of sensitivity mentioned can be obtained in isotopic procedures.

CHEMICAL REACTION OF PRODUCT OR SUBSTRATE

The labeled reaction product may be removed from the reaction mixture or, conversely, the labeled substrate may be removed. This method of separation has been applied in a number of determinations, as in the examples shown at the top of the next column.

In the first reaction, which is used in the determination of choline acetyl transferase, the labeled acetylcholine (enzyme product) is coprecipitated from the mixture with excess choline by ammonium reineckate.[16] To decrease the contamination of the product, an amount of unlabeled CoA about 200 times more than the amount of labeled substrate is added. The isolated precipitate is washed with hydrochloric acid, dissolved in acetone, and counted.

The reagent blank obtained with this procedure is generally only a few counts per minute above what is referred to above as the "equipment blank." Since the substrate used has a specific activity of 10 mc/mmole, the limiting

sensitivity, both theoretical and practical, is 10^{-12} mole. The procedure is convenient and highly reproducible, giving a 3% coefficient of variation.

The procedure described above is used routinely[16] for tissue samples of 0.1 to 20 μg of dry weight. However, in collaboration with Dr. Ezio Giacobini of the Karolinska Institutet (Stockholm, Sweden), we have obtained some preliminary data with this procedure on single cells and, in fact, on parts of single cells.

We thought that the same procedure (precipitation of enzyme product with reinecke salt) would work rather nicely for the measurement of carnitine acetyl transferase, the second reaction, but the reineckate of acetylcarnitine proved to be extremely soluble. However, if I_2–KI is added, the ^{14}C-acetylcarnitine in the presence of substrate amounts of unlabeled carnitine can be isolated quantiatively as a periodide.[17] There is very little reagent blank since the acetyl CoA does not readily contaminate the periodide precipitate.

The third example shows the reaction catalyzed by choline phosphokinase, and a method in which the substrate (^{14}C-choline) is eliminated from the reaction mixture is used for its measurement.[18] The substrates are ^{14}C-choline and ATP, and the labeled enzymatic product is ^{14}C-choline phosphate. After the incubation, sodium hydroxide and then ammonium reineckate are added to the reaction mixture to precipitate ^{14}C-choline; ^{14}C-choline phosphate remains in solution. An aliquot of the supernatant is subjected to another reineckation step in the presence of added carrier choline. The count in the final supernatant due to the residual ^{14}C-choline is thus reduced to zero, i.e., to essentially that of the equipment blank, which is 20 to 25 counts/min. This is a highly reproducible procedure with a coefficient of variation of about 3%. The limiting sensitivity, since we use choline with a specific activity of only 0.75 mc/mmole, is 10^{-11} mole, but with the 20- to 30-mc/mmole labeled choline now available, the sensitivity should be proportionately greater.

The elimination of the substrate is also the most effective way of measuring the products of two other enzyme-catalyzed reactions: ethanolamine phosphokinase* and N-methyltransferase.[19] In each case the labeled substrate is removed after the incubation, leaving behind the labeled product of the enzyme reaction.

SOLVENT EXTRACTION

The essential feature of this method is an extraction system in which recovery of the labeled product of the enzyme reaction is quantitative but that of the labeled substrate is essentially zero. Thus the reagent blank is zero or equal to the equipment blank.

Three different primary amines may be used as the substrate in determining monoamine oxidase,

^{14}C-serotonin → ^{14}C-aldehyde → (acid)
^{14}C-tyramine → ^{14}C-aldehyde → (acid)
^{14}C-dopamine → ^{14}C-aldehyde → (acid)

and each of the three has been used to study this reaction in nervous tissue.[20] The primary amine is oxidatively deaminated to yield the corresponding aldehyde as the primary product. In the presence of oxygen and an additional enzyme, aldehyde oxidase, which is present in most tissues, some of the aldehyde is oxidized further to the acid. Because of the secondary reaction, the study of the enzymatic step has in the past been difficult, particularly when either oxygen consumption or the appearance of aldehyde was used to measure the reaction.

This secondary reaction presents no particular problem in the isotopic method, and the isotopic procedure for this enzyme has turned out to be one of our simplest. In the postincubation phase we acidify with hydrochloric acid, and thus the acid group, if present, and the aromatic hydroxyl present on each product are both in the undissociated state; both the aldehyde and acidic products are quantitatively extracted from the ionized amine (substrate) into ethyl acetate. Ethyl acetate is immiscible with the incubation mixture and, conveniently, is present as the upper phase in a two-phase system. An aliquot of this organic layer is removed, washed rapidly with another small volume of hydrochloric acid, placed in a scintillation vial, and counted. The coefficient of variation is quite good—2 to 3%—and the limiting sensitivity for the protocol using labeled serotonin with a specific activity of only 1 mc/mmole is 10^{-11} mole.

The reagent blank in the case of labeled serotonin and tyramine has little practical significance (i.e., essentially zero). Various lots of dopamine from different suppliers have, at

*R. E. McCaman, unpublished results.

times, given problems because of trace contaminants that are labeled and are extracted from an acidified aqueous solution into ethyl acetate. This, of course, is a matter of purity and not a deficiency of the procedure.

Some other protocols in which the product is separated by solvent extraction include the methods for measuring the following enzyme reactions:

Substrate(s)	Labeled product
Cytidine diphospho-^{14}C-choline (diglyceride)	^{14}C-phosphatidylcholine
Cytidine diphospho-^{14}C-ethanolamine (diglyceride)	^{14}C-phosphatidylethanolamine
^{14}C-L-α-glycerolphosphate (oleyl CoA)	^{14}C-phosphatidic acid
^{3}H-norepinephrine (S-adenosylmethionine)	^{3}H-metanephrine
^{14}C-adenosyl methionine (dihydroxybenzoate)	^{14}C-3-methoxy-4-hydroxybenzoate
^{14}C-carnitine (oleyl.CoA)	^{14}C-oleylcarnitine

The first two of these reactions are for enzymes involved in the terminal biosynthetic steps of phosphatidylcholine and phosphatidylethanolamine (phosphocholine–glyceride transferase[18] and phosphoethanolamine–glyceride transferase,[21] respectively). The labeled substrates are the water-soluble nucleotides with the label in the base moiety, and diglyceride is a cosubstrate in both cases. The radioactive products, the phosphatidylcholine and phosphatidylethanolamine, are quantitatively extracted into an organic solvent. In both cases the reagent blank is essentially equivalent to the equipment blank.

The method for the third reaction is one currently being developed.[22] With ^{14}C-L-α-glycerolphosphate and long-chain acyl CoA (i.e., palmityl, oleyl, or linoleyl CoA, labeled diacylglycerolphosphate (phosphatidic acid) is formed; it also is very conveniently separated from the substrate by simple solvent extraction of the phosphatidic acid.

The fourth reaction is that catalyzed by the enzyme catechol-O-methyl transferase. An isotopic procedure for measuring this enzyme was developed by Axelrod, Albers, and Clemente[23] using tritiated norepinephrine and S-adenosylmethionine as the substrates. The labeled metanephrine (product of enzyme reaction) is extracted into a toluene–isoamyl alcohol solvent. We experienced a number of difficulties trying to adapt this procedure to a smaller scale but resolved most of these as shown in the next example. The label-carrying substrate is ^{14}C-S-adenosyl methionine together with dihydroxybenzoic acid.[24] Axelrod and Tomchick[25] had shown the latter to be a substrate for this enzyme. Both the carboxyl and the phenolic hydroxyl groups are undissociated at an acid *p*H. Under these conditions, the O-methyl derivative is readily extracted into an organic solvent, e.g., ethyl acetate. Since the ^{14}C-S-adenosylmethionine is not extracted, the blank is very low and recovery of the O-methyl derivative is quantitative. The overall gain is approximately 1000-fold over the isotopic method of Alexrod et al.

The last reaction in this category is that catalyzed by carnitine acyl transferase. The enzyme transfers long-chain acyl groups (i.e., palmityl or oleyl) from the corresponding acyl CoA to produce a long-chain acyl carnitine. When ^{14}C-carnitine is used as the labeled substrate, separation of the substrate and labeled product is complete;[26] oleylcarnitine is readily extracted into an organic solvent system; the labeled substrate (carnitine) is not extracted.

MICRODIFFUSION

The labeled reaction product of the following reactions may be separated by diffusion procedures:

$$^{14}\text{C-5-OH-tryptophan} \rightarrow {}^{14}CO_2$$

$$^{14}\text{C-glutamate} \rightarrow {}^{14}CO_2$$

$$^{14}\text{C-6-phosphogluconate (NADP)} \rightarrow {}^{14}CO_2$$

$$^{14}\text{C-acetate (choline, CoA, ATP)} \rightarrow {}^{14}\text{C acetylcholine}$$

The first three of the above reactions lead to the evolution of $^{14}CO_2$, and the techniques used for them are generally similar. It should be emphasized that these procedures are applicable to the study of all decarboxylation reactions for which the appropriate carboxy-labeled substrate can be obtained or synthesized.

In determining 5-hydroxytryptophan decarboxylase, we use ^{14}C-carboxy-labeled 5-hydroxytryptophan as the substrate.[20] During the reaction the $^{14}CO_2$ generated is trapped in a "ringlet" of base as shown in part *a* of Fig. II-12. The tubes used for these experiments are 2 to 2.5 mm in inside diameter, and the capillary attraction is such that a liquid phase of base can be maintained above the actual incubation mixture during the reaction. After the incubation the tubes are removed from the 38°C bath and put in an ice bath. Another ringlet, of

sulfuric acid this time, is formed in the top of the tube, and the tube is connected by a small piece of polyethylene tubing to another tube containing Hyamine.* The first reaction tube, while connected to the second, is mixed in such a way

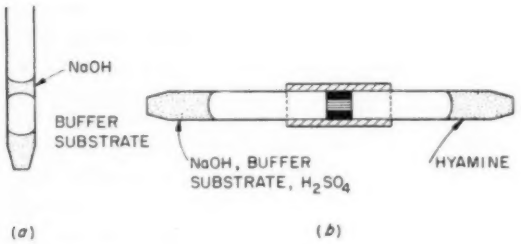


Fig. II-12 Equipment arrangement for the isotopic determination of 5-hydroxytryptophan decarboxylase activity.

that the sodium hydroxide containing the trapped $^{14}CO_2$ and the ringlet of sulfuric acid are both brought down into the bottom of the first tube. The $^{14}CO_2$ gas formed is now free to diffuse over into the second tube, where it is trapped in the Hyamine. An aliquot of the Hyamine is counted. This procedure is very similar to that described for the determination of glutamic decarboxylase developed by Albers and Brady.[27] As far as I know, their measurement of glutamic decarboxylase was the first application of isotopes in quantitative microchemical studies.

In the procedure[28] published recently by Pastan et al., 6-phosphogluconic dehydrogenase activity is measured as a function of the rate of $^{14}CO_2$ evolution from the carboxy-labeled 6-phosphogluconate in the presence of NADP.

It may also be appropriate to include in this category a recently published procedure for the microdetermination of choline acetylase.[29] In this procedure the labeled substrate is ^{14}C-acetate, and the incubation mixture includes choline, CoA, and an acetyl CoA generating system. After the reaction has been terminated and the mixture acidified, the sample is taken to dryness. The ^{14}C-acetic acid volatilizes during this step. The evaporation may be carried out again with carrier acetic acid being added the second time. Thus the nonvolatile labeled material is equivalent to the enzymatically synthesized ^{14}C-acetylcholine. This is an example, again, of getting rid of the substrate rather than isolating the product of the enzyme reaction.

*Hyamine is a commercially available (Rohm & Haas Co.) long-chain base found especially useful in scintillation-counting procedures as a trapping agent for CO_2 and as a solubilizing agent for biological tissues.

Increase of Sensitivity

Let us now briefly consider ways in which the sensitivity of isotopic procedures may be increased so that they could be used in a study of single-cell chemistry. One possible way is the use of labeled substrates with higher specific activities. The theoretical maximum is related to multiples of that obtained for pure ^{14}C, i.e., 64 mc/mmole. However, there are practical limitations to this approach because of blanks, stability, etc., and we probably should assume an upper practical limit of 100 mc/mmole to the specific activity of the substrate, or a limiting sensitivity of 10^{-13} mole for the method.

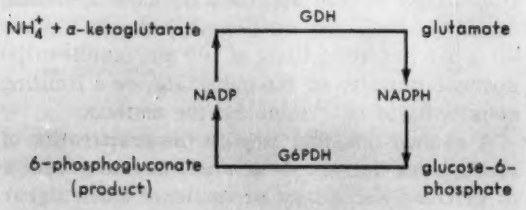
A second possible way is the preparation of labeled derivatives of labeled enzyme products to produce secondary products of much higher specific activity. In the above reaction for ethanolamine phosphokinase, for example, after the enzyme reaction is complete, both the labeled ethanolamine and ethanolamine phosphate may be methylated with labeled methyl groups to give choline and choline phosphate, respectively. Similarly, labeled dinitrofluorobenzene or labeled dinitrophenylhydrazine could be used for increasing the specific activity of enzymatically formed amines and aldehydes, respectively. Such procedures might increase the limiting sensitivity to 10^{-13} or 10^{-14} mole.

Technically there is no reason that isotopes could not be used in conjunction with cycling procedures like those of Lowry and Passonneau,[30] shown in Fig. II-13 for NADP (NADPH) measurements. At the completion of the cycle, the 6-phosphogluconate is measured by the reduction of NADP in the presence of 6-phosphogluconic dehydrogenase. It could just as well be measured by the evolution of $^{14}CO_2$, which would be formed from the substrate 1-^{14}C-glucose-6-phosphate. If isotopic techniques are to offer real utility, however, it would be in facilitating the development of cycles involving substrates and coenzymes other than those already being studied via the pyridine nucleotides. The methods for the enzyme reactions cited above involving acetyl CoA, S-adenosylmethionine, and the nucleotide triphosphates may be adapted to facilitate such procedures.

Principle: A set of conditions is established such that a few molecules of a given substance (limiting substrate) may act catalytically in regulating the production of several thousand molecules of another substance (measured product).

Conditions: Limiting substrate is NADP (NADPH in tissue is selectively destroyed). Auxiliary substrates are glucose-6-phosphate, ketoglutarate, ammonium ions. Auxiliary enzymes are glucose-6-phosphate dehydrogenase, glutamic dehydrogenase.

Cycle Reaction: In the presence of the two enzymes and auxiliary substrates, a few molecules of NADP are continually reduced and oxidized (cycled), producing an equivalent amount of 6-phosphogluconate each time the cycle is completed; the rate (No. of cycles in unit time), as well as the cumulative amount of 6-phosphogluconate, is thus proportional to the amount of NADP originally present:



Measurement: After a fixed time the cycle is stopped by denaturing the enzymes (acid), and the NADP and NADPH are selectively destroyed; the 6-phosphogluconate generated by the cycle is measured by an additional step:

6-phosphogluconate (product) + NADP

$$\xrightarrow{\text{6-PGDH}} \text{ribulose-5-P} + \text{NADPH} + CO_2$$

The amount of NADPH formed here is proportional to the amount of 6-phosphogluconate generated by the cycle, which in turn is proportional to the amount of NADP originally present in the tissue.

Fig. II-13 Cycling procedure for measuring endogenous tissue level of NADP.

Gamma Activation Analysis of Rocks and Ores*

A survey of USSR work on gamma activation analysis indicates this to be a very promising technique for analyzing rocks and ores. Samples to be analyzed are irradiated in the bremsstrahlung beam produced by a betatron, and the induced activities are determined. The use of bremsstrahlung of different energies and selective recording of the activities permit rapid analysis of very complex materials.

This paper discusses the use of gamma activation for analyzing minerals and their derivatives formed in ore beneficiation. The radiation used to activate the elements is the bremsstrahlung produced by the impingement of accelerated electrons on a metal target. The method is faster than neutron activation analysis as usually applied and can determine many elements that are not activated by neutrons. A selective gamma activation method can be used to identify individual elements or groups of elements. In combination with standard programmed methods of data analysis, gamma activation analysis will considerably facilitate mineral composition study.

Modern industrial development is characterized by the requirement for steadily increasing efficiency in analytical methods. Charged-particle accelerators, as generators of neutrons, protons, and gamma radiation, for example, offer wide possibilities for developing analytical methods. Among these is gamma activation, in which a substance is irradiated with high-energy bremsstrahlung and the induced radioactivities, produced by various photonuclear reactions, are measured.

The gamma activation method, proposed[31] in 1954 for determining oxygen in metals and organic substances, has been used for determining impurities in metals or other substances of comparatively simple composition, e.g., oxygen in beryllium, aluminum, titanium, semiconducting materials, and polymers; nitrogen in beryllium, aluminum, and zirconium; carbon in aluminum, beryllium, and iron; and fluorine and chlorine in polymers.[32–38] Gamma activation methods have also been investigated for determining carbon, fluorine, nitrogen, phosphorus, and sulfur in lithium, sodium, and other light metals; zirconium in hafnium; silver in bismuth; titanium in iron; nickel in copper; and rare earths and noble metals in mixtures.[39–43] In these studies the source of gamma radiation was bremsstrahlung with maximum energies ranging from 3 to 40 Mev, obtained from electrostatic, linear, and cyclic (betatron and synchrotron) electron accelerators.

Because of the high-intensity bremsstrahlung generated by modern electron accelerators,

*Based on paper SM-68/59 (in Russian; English translation is USAEC Report ORNL-tr-1097 by Martha Gerrard) presented at the IAEA Symposium on Radioisotopic Apparatus in Industry and Geophysics, Warsaw, Oct. 18–22, 1965, by A. K. Berzin, K. F. Kunetsov, V. V. Sulin, V. I. Belov, G. Ch. Vitozhents, Yu. T. Martynov, V. G. Suslov, and S. I. Shornikov of the All-Union Scientific Research Institute for Nuclear Geophysics and Geochemistry, Moscow.

sensitivity of the gamma activation method is high for many elements. For example,[41] with 27-Mev electrons the limits of detection are

Si, W, Pb, Sr	10^{-5} to 10^{-6} g
S, Cr, Mo, Pt, Fe	10^{-6} to 10^{-7} g
C, N, O, F, P, Cl, K, Ti, Cu, Zn, Ag, Ta, Zr	10^{-7} to 10^{-8} g
Mn, Ni, Hf, Sb	10^{-8} to 10^{-9} g
Co, As, Nb, Ca	10^{-9} to 10^{-10} g

With many of these elements in samples of relatively simple composition, the analytical sensitivity[41,42] is decreased by a factor of only about 10 if the products are not separated.

Extensive work has been done in the USSR on gamma activation analysis of rocks and ores based on photoneutron and photoproton reactions, which may be induced by the bremsstrahlung from a relatively simple electron accelerator, the betatron. With a standard laboratory betatron giving 15- to 30-Mev bremsstrahlung, the sensitivity of the method is 10^{-2} to 10^{-3} wt.% for 100-g samples, which is entirely satisfactory for most geological and ore-beneficiation work. The use of gamma activation was proposed in the USSR in 1959 for analyzing ores and in 1960 for determining the elements — e.g., magnesium, silicon, copper, vanadium, and molybdenum — directly in rock stratums traversed by boreholes.[44,45] Further development work has been primarily on analysis of rocks, ores, and substances produced in their processing.[46–51]

Selective Gamma Activation Analysis

The elemental composition of rocks and ores is very complex. Most of the elements can be activated by high-energy gamma radiation, but a complete elemental analysis by simultaneous activation of all elements present may be very difficult. However, successive steps — selective activation of the different elements by bremsstrahlung with different maximum energies and selective recording of the induced activities — may make such an analysis possible. Selective gamma activation is based on the threshold nature of photonuclear reactions and on differences among the energy thresholds and the shapes of the excitation curves for different nuclides (Table II-4).

As an actual analytical problem, let us consider the determination of copper, zinc, barium, and iron in an ore. Samples irradiated by 15.6-Mev bremsstrahlung and cooled long enough for essentially complete decay of the short-lived aluminum and lead show five activities: copper, zinc, barium, silicon, and iron. With 13.8-Mev bremsstrahlung, iron is not activated. With 12.8-Mev bremsstrahlung, the copper, zinc, and barium are detected. Barium and copper appear with 11.6-Mev irradiation and barium alone with 10.8-Mev irradiation. In each case the induced activities may be differentiated by the differences in the half-lives of the activation products and the natures and energies of their radiations. Similar procedures (Table II-5) may

Table II-4 GAMMA ACTIVATION PROPERTIES OF ELEMENTS IN ROCKS AND ORES*

Activated isotope	Natural abundance, %	Reaction	Energy, Mev		Max. reaction cross-section, barns	Radioactive product of reaction	Half-life	Type of decay	Energy, Mev	
			Threshold	Max.					Particles	Quanta
^{12}C	98.89	(γ,n)	18.6	22.8	10.4	^{11}C	20.5 min	β^+	0.96	0.511
^{14}N	99.64	(γ,n)	10.5	22.5	2.8	^{13}N	10.0 min	β^+	1.19	0.511
^{16}O	99.76	(γ,n)	15.6	21.9	14.0	^{15}O	2.03 min	β^+	1.73	0.511
^{19}F	100.0	(γ,n)	10.4	22.2	3.5	^{18}F	1.87 hr	β^+	0.65	0.511
^{24}Mg	78.6	(γ,n)	16.5	19.4	9.8	^{23}Mg	12 sec	β^+	3.0	0.511, 0.44
^{26}Mg	11.29	(γ,p)	14.3	22.0	25.0	^{25}Na	60 sec	β^-	3.8 2.8	0.4–1.61
^{63}Cu	69.1	(γ,n)	10.8	17.5	11.0	^{62}Cu	9.8 min	β^+	2.91	0.511, 0.66–2.24
^{64}Zn	48.89	(γ,n)	11.6	18.7	120	^{63}Zn	38 min	β^+	2.36	0.511, 0.67–2.9
^{90}Zr	57.46	(γ,n)	12.37	18.0	150	^{89m}Zr	4.4 min	I.T.	None	0.59–1.5
^{138}Ba	71.86	(γ,n)	8.51	No data		^{137m}Ba	2.6 min	I.T.	None	0.661
^{204}Pb	1.48	(γ,n)	8.7	22.0	280	^{203m}Pb	6 sec	I.T.	None	0.83
^{208}Pb	52.3	(γ,n)	10.8	14.0	30.0	^{207m}Pb	0.8 sec	I.T.	None	0.57–1.06

*Only those elements found in amounts greater than hundredths of a percent are included.

be used for analyzing, for example, titanium–zirconium ores for titanium and zirconium with separation of the titanium, zirconium, and silicon activities; phosphorites for phosphorus, fluorine, silicon, and potassium; potassium "salt" (a potassium fertilizer largely KCl) for potassium, with separation of potassium and chlorine; and coal for carbon, oxygen, and silicon, with separation of carbon, oxygen, silicon, and potassium.

Table II-5 CHARACTERISTICS OF GAMMA ACTIVATION ANALYSIS METHODS FOR ROCK AND ORE SAMPLES

Sample	Element determined	Recommended max. betatron energy, Mev	Sensitivity, wt.%	Analytical precision, %	Time for analysis of one sample, min
Ti–Zr ores and concentrates	Zr	13.8	7.5×10^{-3}	5–10	10
	Ti	14.2	2×10^{-1}	5–10	20
Phosphorites, apatites	P	15.6	10^{-2}	10–15	20
	F	15.6	10^{-2}	10	30
Polymetal and Cu ores	Cu	13.8	1.2×10^{-2}	10	20
	Zn	13.8	2.5×10^{-2}	10	20
	Ba	13.8	1.2×10^{-1}	10	10
Coal	C	28	1×10^{-3}	5–10	20
	O	19	5×10^{-2}	5–10	10
	C + O	23	8×10^{-1}	5	10
High-ash coal	C	28	10^{-2}	30	20
Sedimentary rock	C	28	10^{-1}	20	20
	O	19	5×10^{-2}	5–10	10
	C + O	23	5×10^{-3}	5–10	20

TECHNIQUES

Pulverized samples are irradiated in the bremsstrahlung beam produced by 15- and 30-Mev maximum energy electrons (at rates of 6 and 160 r/min, respectively) and cooled 1 to 3 min before the induced gamma activities are recorded. The maximum electron energies are continuously recorded (14.2 and 28.7 Mev) and are automatically controlled within ±0.1 Mev by stabilizing electrodes. The cylindrical sample containers are made from Plexiglas or from a vinyl material with Plexiglas bottoms and covers. Most of the containers are 60 mm in inside diameter and 50 mm deep and have 2-mm-thick bottoms and covers.

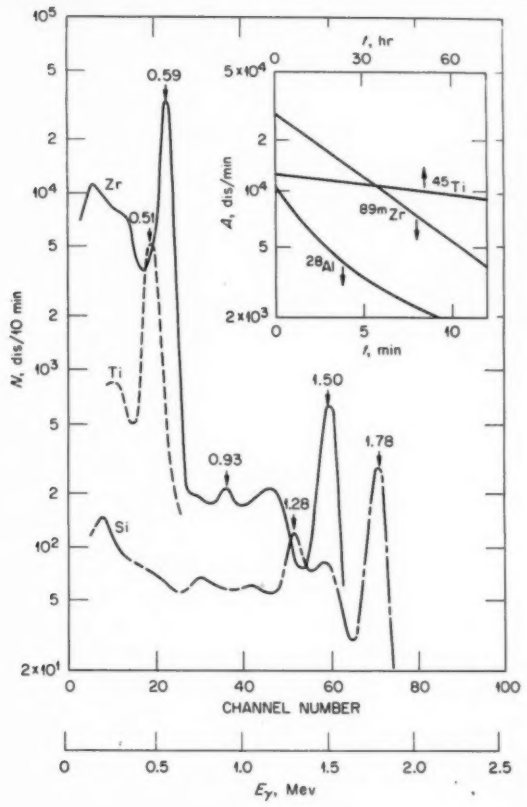
Because of the high penetrating power of hard bremsstrahlung radiation, samples weighing as much as 100 to 200 g are analyzed; the pneumatic tubes used to carry samples in neutron activation analysis, where weights are milligrams or grams, therefore cannot be used. In the mechanical conveyer system used in gamma activation analysis at the All-Union Institute, the samples are moved by a manipulator from the storage bin, which has a capacity of five 100-mm-long cassettes, to the conveyer system. A magnetic device and cable arrangement move them along a monorail to the betatron, then to the measuring apparatus, and eventually back to the storage bin. The rate of transfer of the cassette with the irradiated sample is adjustable to a maximum of 2 m/sec for loads of 100 to 700 g. About 0.5 sec is required for the manipulator to move the irradiated containers to the storage bin.

The bremsstrahlung-induced activities are measured with the samples in the same container in which they were irradiated except when the irradiated cassette is a source of interfering activity or self-absorption effects must be excluded. In such cases special measuring containers[47] are used. Activities are recorded by a spectrometer, with a NaI(Tl) crystal of 70 or 40 mm diameter and a photomultiplier photocathode of 75 or 45 mm diameter. Pulse amplitudes are determined with 50-, 100-, or 256-channel analyzers.

RESULTS

A typical set of measurements is shown graphically in Fig. II-14. (The original paper shows 10 such sets.)

(Martha Gerrard)



Photopeak	Reaction
0.511 Mev	$^{46}Ti(\gamma,n)^{45}Ti$
0.590 Mev	$^{90}Zr(\gamma,n)^{89m}Zr$
0.930 Mev	$^{90}Zr(\gamma,n)^{89}Zr$
1.28 Mev	$^{30}Si(\gamma,p)^{29}Al$
1.50 Mev	$^{90}Zr(\gamma,n)^{89m}Zr$
1.78 Mev	$^{29}Si(\gamma,p)^{28}Al$

Fig. II-14 Gamma spectrums and decay curves for activation products of principal elements in titanium–zirconium ores. $E_{max.}$ *= 13.8 Mev,* $t_{irrad.}$ *= 15 min,* t_{decay} *= 1 min.*

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Isotopic Method for Determining Enzyme Activity in Microgram and Submicrogram Quantities of Tissue

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Section

III

Isotopes and Radiation Technology

Food Irradiation

The first article in this section describes the irradiators that the U. S. Atomic Energy Commission (AEC) has built or is building for the food-irradiation program. The second article summarizes hearings on the radiation processing of food held on June 9–10, 1965, before the Subcommittee on Research, Development, and Radiation of the Congressional Joint Committee on Atomic Energy. The programs of the AEC on pasteurization of foods, of the Army on sterilization of foods, and of various industrial firms and other government agencies are summarized. The third article reviews the literature and status of work on the quality and wholesomeness of radiation-pasteurized marine products, particularly fish fillets.

AEC Food-Irradiator Program[1]

The irradiation facilities that are being built in the AEC food-irradiation program are described. These irradiators, which have capacities ranging from that needed for university research to that required for semicommercial operations, use ^{60}Co or ^{137}Cs as a source of radiation. Essential characteristics such as source strength, production capacity, radiation dose range, and cost are given for the research irradiators, Marine Products Development Irradiator, Grain Products Irradiator, Mobile Gamma Irradiator, On-Board Ship Irradiator, the Hawaii Development Irradiator, and the Portable Irradiator.

One of the major portions of the AEC program for developing commercial food-irradiation techniques is the design and construction of a group of irradiation facilities that can be used in the food-irradiation program from laboratory to commercial scale. These irradiators, which were reviewed briefly in a past issue of *Isotopes and Radiation Technology*,[2] are listed in Table III-1.

The technology of source and irradiator design has been developed concurrently with food-irradiation processes. Prior to 1961, when the first 35,000-curie ^{60}Co research irradiator was installed at the Massachusetts Institute of Technology (MIT), no comparable irradiator existed to perform the services required. Food-irradiation research has been and is forcing the development of moderate- and large-scale ^{60}Co irradiators and is helping to pave the way for commercial isotopic radiation applications in other fields such as medical-supply sterilization.

Below, after a brief discussion of source selection, the design and operation of each of the irradiators are discussed. The order of discussion is roughly chronological, i.e., the research irradiators, which are the oldest and most used irradiators, are discussed first, and the Portable Irradiator, which is still in the design stage, is discussed last.

Source Selection

The selection of ^{60}Co as the radiation source for initial development by the AEC was based on many factors: It offered no induced activity problems when used for food processing; it was available in adequate quantities as a reactor by-product; its gamma rays were highly penetrating and therefore applicable to bulk irradiation; technology associated with source fabrication and encapsulation was better developed than for other source possibilities; and, its cost, already reasonable, could be expected to decrease considerably.

Today a number of radiation sources are either available or are being developed to meet process irradiation requirements, including those for food irradiation. High-energy-electron source manufacturers, who early in the food program contributed immeasurably to the initial development of irradiation technology, now offer a variety of electron generators ap-

Table III-1 AEC FOOD IRRADIATORS*

Irradiator	Purpose	Location	Status	Design	Construction
Research irradiators	Immediate irradiation service for contractors in food-irradiation program	MIT	Operating	BNL	Processing Equipment Corp., Lodi, N. J.
		U. of Calif. (Davis)	Operating		
		U. of Wash.	Operating		
		U. of Fla.	Operating		
		LSU	Operating		
		U. of Hawaii	Operating (source only provided by AEC)		
Marine Products Development Irradiator	Semicommercial seafood irradiation	U. S. Dept. of Interior, Technological Lab., Gloucester, Mass.	Operating	Associated Nucleonics, Inc.,† Garden City, N. Y.	Associated Nucleonics, Inc.
Grain Products Irradiator	Disinfestation of bulk grain or packaged products	U. S. Dept. of Agriculture, Entomological Research Center, Savannah, Ga.	Completion of construction was scheduled for July 1966	BNL and Vitro Engineering Co., New York	Lockheed-Georgia Co., Marietta, Ga.
Mobile Gamma Irradiator	Demonstration of feasibility of irradiation processing of fruits at harvest locations	U. of Calif. (Davis) headquarters	Operational June 1966	Vitro Engineering Co.	Lockheed-Georgia Co. Marietta, Ga.
		U. S. Dept. of Interior, Technological Lab., Gloucester, Mass.			
On-Board-Ship Irradiator (3)	Research support at sea for seafood-irradiation program		Operating	BNL	Nuclear Materials & Equipment Corp., Apollo, Pa.
		LSU, operator, Pascagoula, Miss. Headquarters	Completion of construction was scheduled for June 1966	BNL	Radiation Facilities, Inc., Lodi, N. J.
	Cooperative international food program	On loan to Israel	Operating	BNL	Nuclear Materials & Equipment Corp.
Hawaii Development Irradiator	Semicommercial irradiation processing of tropical fruit	U. of Hawaii, Honolulu	Design (completion early 1967)	Nuclear Materials & Equipment Corp.	Nuclear Materials & Equipment Corp.
Portable Irradiator	Research support for food-processing industries	At company site	Under construction (completion 1966)	Vitro Engineering Co.	Not selected

*All have ^{60}Co sources except Portable Irradiator which uses ^{137}Cs.
†Now Associated Engineers and Consultants, Inc.

plicable to irradiation processing of foods.* The versatility and dependability of these units have undergone constant improvement, and many are currently used in commercial processing. The Food and Drug Administration (FDA) has approved the use of electron beams for sterilization of bacon[3] and disinfestation of wheat and wheat products.[4]

The conversion of electrons to X rays or bremsstrahlung for radiation processing also appears quite feasible, primarily in the intermediate-energy-electron range. At least one electron-machine manufacturer has revealed plans for such a unit. Although widely demonstrated at the laboratory level or manufacturer's plant, such X-ray machines have as yet not been applied to commercial processing.

Several radioisotopes hold a good potential for commercial radiation processing, but for food irradiation ^{60}Co stands out as especially well-suited. A program recently started at Brookhaven National Laboratory (BNL) on the evaluation of ^{137}Cs as a radiation source for food preservation is expected to put this isotope in the same category.

With these several sources available for commercial processing, the inevitable problem of source selection arises. Unfortunately no stock answer or measurement can be applied. A thorough evaluation of each radiation source applied to specific products under desired processing conditions is the only way in which an intelligent comparison can be made. Such an evaluation, to be meaningful, should encompass all elements of the potential irradiation process from capital costs to product handling. A number of engineering firms are now qualified to make such an evaluation.

*See, for example, the feature article in this issue, "Particle Accelerator Uses for Industry," by E. A. Burrill.

Research Irradiators

Research irradiators[5] were designed principally for installation at research sites to provide immediate irradiation services to contractors doing research in the AEC food-irradiation program. Their characteristics are:

Purpose:	Immediate availability for research support
Type:	Double-plaque source, located in pool
Source:	25,000–50,000 curies of ^{60}Co
Capacity:	75 lb/hr at 1 Mrad
Facility cost:	$35,000 exclusive of source and housing

These irradiators served as the backbone of the program, providing nearby radiation support facilities and obviating the need for artificial preservation of foods during transit to a remote irradiation site. Simplicity, ease of maintenance and operation, and versatility are prime characteristics.

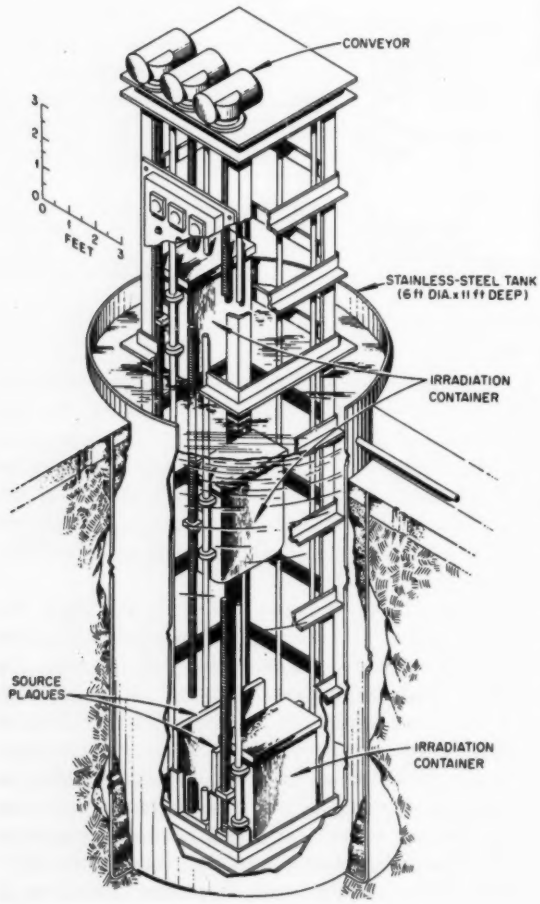

Fig. III-1 Research irradiator.

A typical research irradiator (Fig. III-1) consists of two ^{60}Co source plaques 23 by 25 in. mounted in the bottom of a stainless-steel water-filled tank 11 ft deep and 6 ft in diameter. The material to be irradiated is placed in watertight containers, which can hold packages up to 14 by 18 by 6 in., and is lowered by an elevator to the bottom of the tank. Three containers may be exposed simultaneously, one between the two plaques and one on the outer side of each. The dose received by material in the center container is, of course, about twice that received by the outside containers. One of the three irradiation containers is provided with both temperature and atmosphere control to facilitate experimental work. A central control panel is provided so that a well-trained technician can safely operate the facility. The period of irradiation can be preset and the irradiation performed automatically. Safety instrumentation and a water-purification unit are included.

The research irradiator recently installed at the University of Hawaii[5,6] is slightly modified from the usual one. Rather than having three rectangular box containers, this irradiator has one central rectangular container between the source plaques, but one outside position has three 6-in.-diameter tubes and the other, five 3-in.-diameter tubes (Fig. III-2). This design change facilitates exposures of smaller containers and slightly lowers the construction cost.

Marine Products Development Irradiator

The Marine Products Development Irradiator (MPDI) is a semiproduction facility designed to irradiate quantities of seafoods sufficient for evaluating shipping, storage, and consumer acceptance and for other large-scale tests and to help develop information on the economics of radiation processing. It has been described in detail in an earlier issue of *Isotopes and Radiation Technology*[7] and in several reports;[8,9] its essential characteristics, in summary, are:

Purpose:	Semicommercial seafood irradiation
Type:	Fixed facility; four-pass quadrant irradiation, pool storage for source
Source:	250,000 curies of ^{60}Co
Capacity:	2000 lb/hr at 250,000 rads
Facility cost:	$600,000 exclusive of source

Boxes containing 30 lb of fish fillets are normally irradiated in this facility. These boxes are loaded in pairs onto a conveyor out-

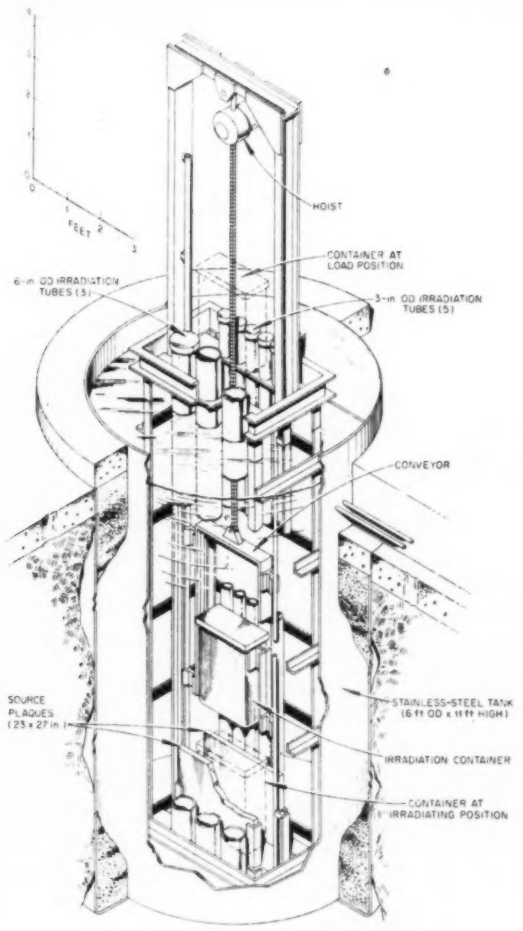

Fig. III-2 Hawaiian Research Irradiator.

side the radiation cell and transported through a labyrinth to an inner conveyor system, which passes them under and over the source (Fig. III-3). The boxes are then brought out of the cell, their positions on the conveyor are reversed, and they are then passed under and over the source again. The ^{60}Co is stored in a pool when not in use and is raised by an elevator when needed.

Grain Products Irradiator

The AEC, in cooperation with the U. S. Department of Agriculture (USDA), has established a program designed to determine the role that radiation might have in wheat and grain preservation. To support this research, the AEC will provide a ^{60}Co unit capable of irradiating bulk wheat as well as packaged products. The unit is to be located at the USDA's Entomological Research Center in Savannah, Ga. The characteristics of this irradiator[10–12] are:

Purpose:	Disinfestation of bulk grain or packaged products
Type:	Fixed grid of ^{60}Co strips; two irradiation positions, one for bulk grain and one for packages
Source:	25,000 curies of ^{60}Co
Capacity:	5000 lb/hr bulk or 2800 lb/hr packaged product, either at 25,000 rads
Facility cost:	$200,000 exclusive of source

The irradiator is in a concrete cell, about 15 ft square by 20 ft high, partially housed in a prefabricated building. The source is 25,000 curies of ^{60}Co in 13 parallel rods in a horizontal plane (Fig. III-4). Elements of higher activity are used in the edge rods and at the ends of the rods to achieve dose uniformity over the entire grid. The source rods can be positioned manually in two different locations, one for bulk grain irradiation and the other for package applications (Fig. III-5).

For bulk irradiations the grain is conveyed to the top of a 4-sq ft bin by mechanical conveyors. From the top of the bin, it is moved past the source grid by gravity and mechanical vibrations. The grain bed completely surrounds the source rods except at the perimeter and extends 3 ft above and below the plane of the grid. This geometry yields a radiation efficiency of almost 50%. The dose is determined by regulating the flow rate. From the bottom of the bin, the grain passes through a rotating feed valve and is pneumatically conveyed to storage.

For package irradiation, the packages are arranged in aluminum boxes, which are mechanically conveyed over and then under the source array. The movement of the boxes is stopped from time to time to achieve the desired dose.

Mobile Gamma Irradiator

The Mobile Gamma Irradiator (MGI) (Fig. III-6) is an irradiation facility mounted on a trailer that can be taken where fruit is being harvested or processed. Associated Nucleonics, Inc., in a preliminary design study, explored the feasibility of transportable ^{60}Co irradiators.[13] Several design alternatives suggested that such a unit, while hampered by weight limitations, could still maintain a re-

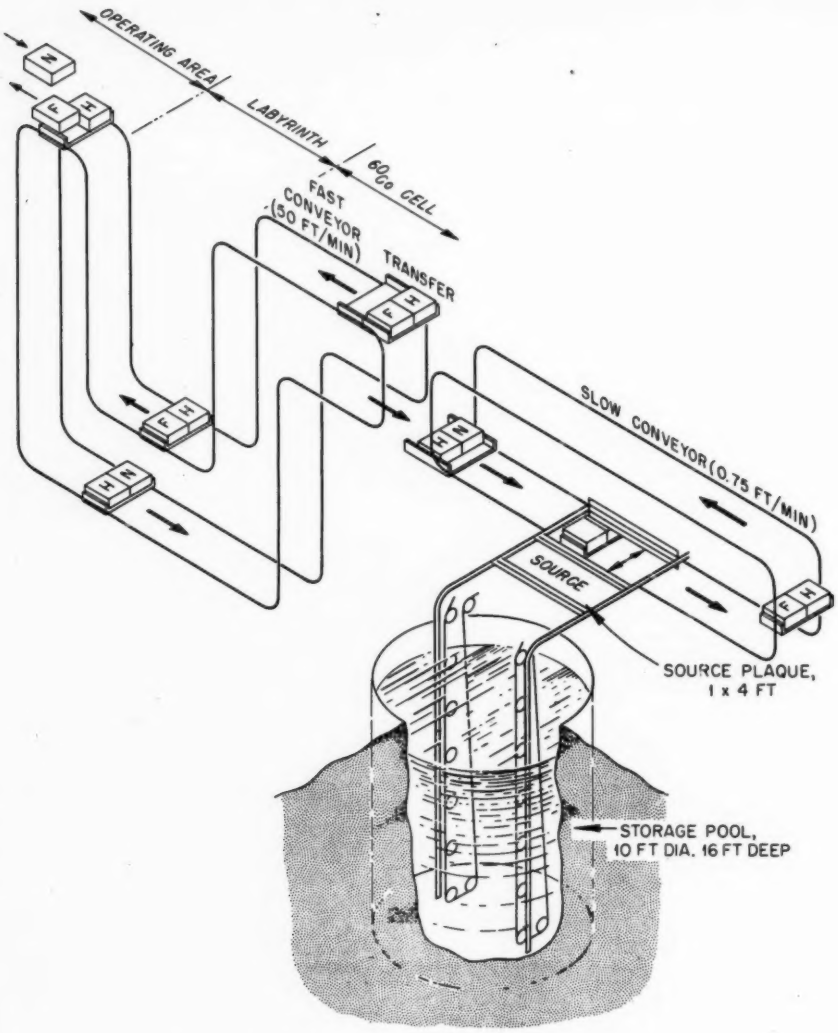

Fig. III-3 Marine Products Development Irradiator.

spectable product throughput and would be especially valuable to demonstrate the applicability of wide-scale food irradiation. Further, it could be used at a number of locations in cooperative industry–AEC demonstration programs, in processing of foods for test marketing and in shipping, storage, and distribution tests. Using this study as a basis, the AEC selected the Vitro Engineering Co., as a result of competitive bids, to detail design and supervise construction of the present mobile unit.

The final design criteria[14] were a compromise between weight requirements, source efficiency, transportability, product throughput, and uniformity of dose. Since initial use will be with fruit products, a basic container size of approximately 9 by 14 by 20 in. (representing a stack of two standard strawberry flats) was selected. This also accommodates the containers used for many other fruits. The characteristics of this irradiator are:

Purpose:	Large-scale demonstration of feasibility of radiation processing of fruits; economics studies
Type:	Mounted on truck, mobile; 59 tons complete, including tractor and trailer; four-pass quadrant irradiation
Source:	100,000 curies of ^{60}Co
Capacity:	1000 lb/hr, 200,000-rad dose
Facility cost:	$300,000 exclusive of source

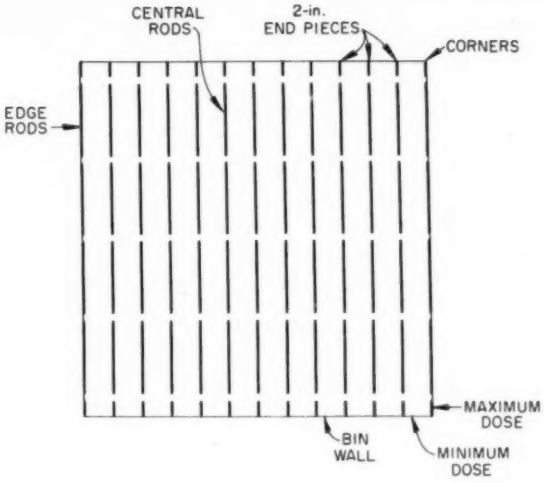

Fig. III-4 Plan of source configuration for Grain Products Irradiator. The rods have the following specific activities: central rods, 5.5 curies/g; edge rods, 12.7 curies/g; 2-in. end piece, 23.1 curies/g; and corners, 35 curies/g.

The MGI consists of the irradiator body, a source cask, hydraulic system, refrigeration and ventilating systems, instrumentation, and conveying system, all mounted on a trailer bed. Both the irradiator body and the cask are welded carbon steel filled with lead.

The source is 19 strips of doubly encapsulated ^{60}Co in a plaque measuring 15 by 18 in. and containing 100 thousand curies. Because of the severe limitations in weight and maximum-to-minimum product-dose ratio, the quadrant irradiation principle is used in which the product completely overlaps the source except at the edges. This, along with the high specific activity of the source (70 to 80 curies/g), minimizes source size and therefore shielding volume and weight.

The products are placed in an aluminum carrier, and they enter the irradiator via a shielded shuttle system. Each container makes a series of controlled movements, passing over and under the horizontal source four times, and finally exiting by the same shuttle. The movements are activated by a series of hydraulic cylinders. A strict maximum-to-minimum dose uniformity ratio of 1.28 is maintained, so that a desired dose of 175 thousand to 225 thousand rads, applicable to several fruits, is obtained.

The refrigeration and ventilating systems maintain the product at temperatures between 32 and 41 °F and remove ozone.

This irradiator will be operated by the group at the Davis campus of the University of Cali-

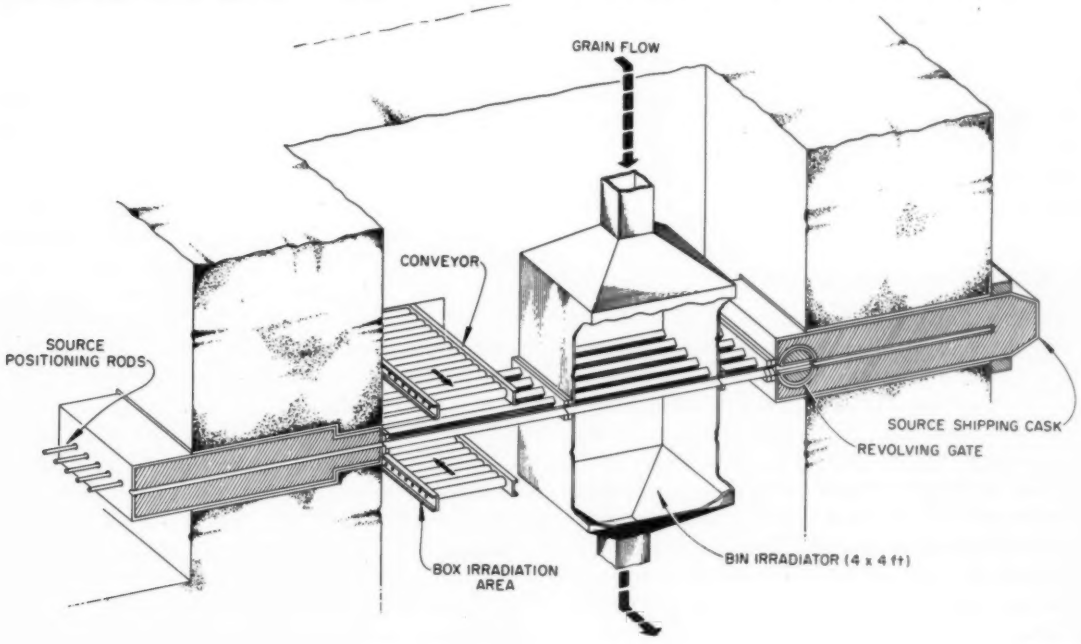

Fig. III-5 Grain Products Irradiator.

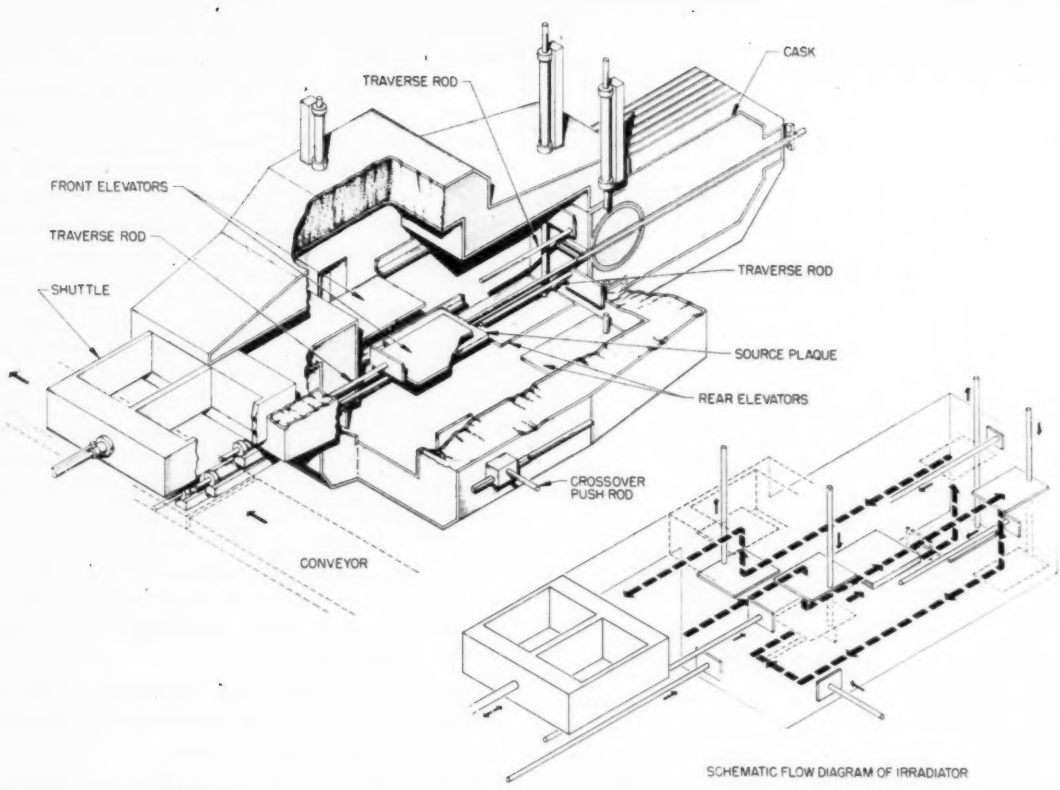

Fig. III-6 Mobile Gamma Irradiator.

fornia that has been doing research on fruit irradiation for the AEC. Irradiation of strawberries will be tested first since this fruit appears to be the prime fruit candidate for radiation processing.

On-Board-Ship Irradiator

Earlier fishery studies have established that irradiation of fish should take place as soon after catch as possible. While a land-based unit such as the MPDI represents the earliest time after landing that irradiation could take place, there is the alternative of irradiation at sea. Bacterial counts on either semiprocessed or unprocessed whole fish could be reduced by the application of a light (100 thousand rad) dose immediately after catch and of a second moderate dose after final processing and packaging.

An earlier study conducted by Vitro Engineering Co.[15] described a large-scale ^{60}Co unit capable of processing 7500 lb of fish per hour with a dose of approximately 100 thousand rads. While this unit is applicable for installation on board larger fishing vessels, its construction by AEC is not anticipated. Rather, to meet the immediate need for processing at sea—where fishing runs often last 1 to 2 weeks—two smaller self-contained units, designed initially at BNL,[5] are now nearing completion or are in operation (Fig. III-7). They will be used by the U. S. Department of the Interior's Technological Laboratories at Gloucester, Mass., and Seattle, Wash., and by Louisiana State University in the latter's shrimp-irradiation program. The characteristics of this irradiator are:

Purpose:	Research support for seafood-irradiation program
Type:	Transportable, 17-ton self-contained unit
Source:	30,000 curies of ^{60}Co
Capacity:	150 lb/hr at 100,000 rads; continuous pasteurization of seawater for storage purposes
Facility cost:	$36,000 exclusive of source

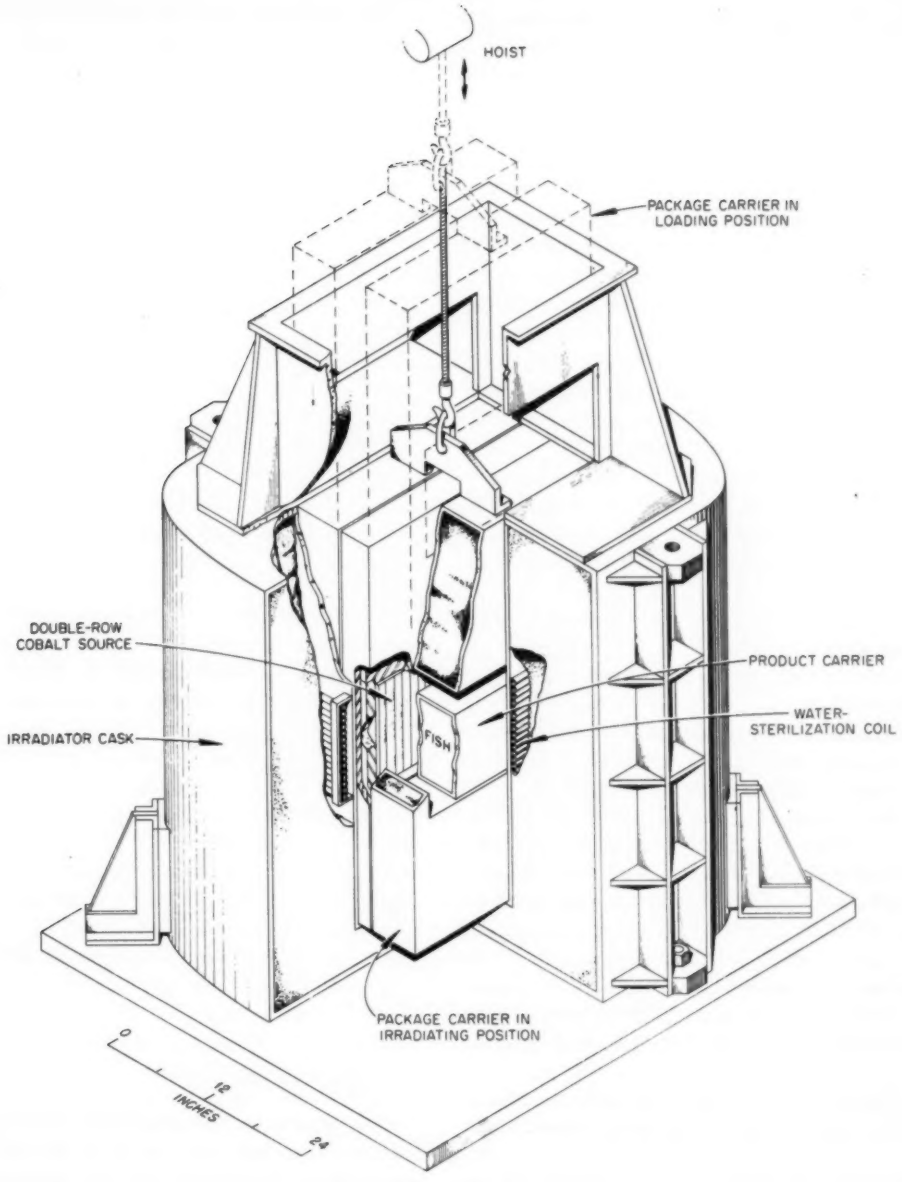

Fig. III-7 On-Board-Ship Irradiator.

This irradiator stands on a base 4 by 5 ft, and, when assembled and ready for operation, it is about 8 ft high. The 30-thousand-curie ^{60}Co source is a plaque 18.5 by 26.5 in. The holder for this plaque is attached to a lead plug, and the entire assembly is bolted in place so it cannot be removed except at a suitable radiation cell or pool facility. The shielding is 11 in. of lead and 1 in. of steel.

Two boxes, each 15 by 19 by 16 in., can be introduced into the irradiation chamber simultaneously through the manually loaded shielded shuttle on top. Boxes must be exposed on each side of the plaque to achieve uniform irradiation.

In addition to the throughput of 150 lb of fish per hour, the irradiator also has the concurrent capability of pasteurizing refrigerated

seawater for experimental use in keeping the catch fresh while on the ship. Preliminary work has shown this method of maintaining fresh quality to be quite feasible.

Hawaii Development Irradiator

The design work on the Hawaii Development Irradiator, to be located in Honolulu, was started in the fall of 1965 by Nuclear Materials & Equipment Corp.[16] This irradiator will be used on a semicommercial scale to study radiation preservation of tropical fruits for shelf-life extension, spoilage reduction, and quarantine control. The state of Hawaii is investing about $180,000 in the building to house the irradiator and has authorized the state Department of Agriculture to begin a large-scale development plan for use of the irradiator in commercialization ventures with the Hawaiian fruit industry.

The basic design features established for this irradiator are:

1. Source: ^{60}Co, double encapsulated using BNL standard strips.

2. Irradiation target: primary design emphasis on papayas, the most likely principal product, with mangoes the second choice.

3. Product used for design basis: papayas, prepacked (after hot- and cold-water dipping) in "standard" packages having external dimensions of 17 by 12 by 7.5 in. and internal dimensions of 16.5 by 11.5 by 6.5 in. and containing 10 lb of fruit.

4. Design throughput rate: mean throughput rate of 4000 lb of papayas per hour for first 3 years of operation based on 24 hr/day operation.

5. Design dose for papaya: 75,000-rad minimum dose; maximum-to-minimum dose ratio of 1.30.

6. Feed and transport system: manual initial loading and final unloading of packages, with all other operations automatic and packages transported by removable carriers capable of handling 6000 lb/hr. Normal attitude of packages to remain the same during the irradiation cycle. Variable-speed sections may be used to minimize package inventory and total time within the irradiator.

7. Source storage: pool system, provided with clarifier, filter, demineralizer, lighting, and maintenance tools. Shipping cask for ^{60}Co is not included as an irradiator component.

8. Irradiator temperature control: no special controls to be provided, operation to be substantially at the 70°F temperature maintained by the air-conditioning system of the irradiator building. Initial product feed temperature may be lower than 70°F when put into the irradiator transport carriers as the result of refrigerated storage.

9. Air sweep through irradiator chamber and labyrinth: adequate to hold ozone content of air discharged from the irradiator at 4 ppm maximum.

10. Dose range: 20 thousand to 500 thousand rads for continuous operations. Higher doses to be obtainable by incorporation in the conveyor system of a cycle timer with override switching device.

11. Shielding: normal reinforced concrete.

12. Access to storage pool: overhead "stepped" removable reinforced-concrete roof plug.

13. Irradiator integration into facility building: irradiator placed at one end of processing building to provide flexibility and future additions or modifications to the building.

The characteristics thus established for this irradiator are:

Purpose:	Semicommercial scale processing of tropical fruits for large-scale shipping, storage, distribution, test marketing, and economic studies
Type:	Fixed facility; pool storage for source
Source:	250,000 curies of ^{60}Co
Capacity:	4000 lb/hr at 75,000 rads
Cost:	AEC: $300,000 plus source; state of Hawaii: $180,000

Portable Irradiator

The AEC has had numerous expressions of interest from the food industry related to product testing, preferably at the individual company site. Companies, in effect, want to prove to themselves on their home territory that irradiation preservation can be applicable to their products and their methods of processing.

To meet these requests, the AEC has recently authorized the Vitro Engineering Co. to design a small-scale unit, probably to be mounted on a flat-bed trailer, which could be transported to various companies for their own end-product testing. Although it is premature to specify firm operating characteristics, the tentative characteristics of this irradiator are:

Purpose:	On-site irradiation at company location
Type:	Portable, trailer-mounted unit; irradiator weight, 15 to 20 tons
Source:	^{137}Cs; 100,000 to 150,000 curies
Throughput:	100 lb/hr at 200,000 rads
Cost:	$75,000, including ^{137}Cs

Completion of the unit is expected by September or October of 1966.

(Robert H. Lafferty, Jr.)

1965 Joint Committee Food-Irradiation Hearings

A full summary of hearings on irradiation processing of food, held by the Joint Committee on Atomic Energy on June 9–10, 1965, is presented. Testimony of 20 witnesses from industry, AEC, and the Department of the Army is given, with some of the questions and answers that followed formal testimony. General consensus is that the Army and AEC will continue to support various programs for at least 5 more years. Industrial representatives felt that commercialization of the process will depend directly on FDA approval.

Hearings before the Subcommittee on Research, Development, and Radiation of the Joint Committee on Atomic Energy of the Congress of the United States were held June 9–10, 1965, on irradiation processing of foods. These proceedings are now available.[17] At the time of the hearings, reports appeared in many trade magazines giving brief impressions of the hearings.[18–21] The published hearings are an excellent reference source. Many of the witnesses had bibliographies with their formal, written testimony. A large amount of material was submitted as backup for the record; exactly how much is evident from the fact that the actual hearings cover only 210 pages of the volume's 821 pages. Appendix 15 is an annotated bibliography[22] containing 300 entries on "Wholesomeness of Irradiated Foods." The following review is a full summary of the hearings, reported here as part of the continuing extensive coverage of this subject by *Isotopes and Radiation Technology*, [1: 18-21, 181-187, 245-255, 310-324(1963–1964); 2: 50-54, 264, 317(1964–1965); 3: 30-48(1965).]

Titles of the individual speeches are provided by the editor to help the reader locate topics of specific interest.

The hearings began with opening remarks by Rep. Melvin Price, chairman of the Subcommittee on Research, Development, and Radiation, who was presiding. He stated that the Joint Committee on Atomic Energy (JCAE) regards irradiation processing of foods as "a very important peaceful use of the atom," (1)* and that the purpose of the hearings was "to receive current information on the achievements of the program and hear of present and future plans for the governmental agencies and private industry as well." (1) The last previous hearings on this subject were held March 1962 and May 1963. A milestone was achieved in February 1963 when the FDA approved irradiated bacon for public consumption. He stated, "In my view, the next significant milestone that lies ahead is the achievement of what might be termed 'commercialization' of the food irradiation process, including operation of a production-size irradiation facility under conditions normal to the food industry." (2)

With that introduction, testimony began.

History and Overall Review of AEC Program

By Commissioner James T. Rameyt

Commissioner Ramey opened his testimony with a short history of the food-irradiation program. He pointed out that the division between the use of radiation sterilization by the Army and the use of radiation pasteurization by the AEC dated from the January 1960 hearings. The intention of the Commission then, as now, was the establishment of technical and marketing feasibility for using low-dose radiation to process perishable foods, including safety clearances from the FDA.

The initial study involved five types each of seafood and fruit. The seafoods remain promising, but peaches and grapes have been dropped from the fruit list. Two new fields of interest are the inhibition of sprouting in potatoes and the disinfestation of grain, cereal products, and certain fruits. Program emphasis is changing to include activities designed to encourage early commercialization of the technology. There will be cooperative projects with industry, such as pilot facilities. Commissioner Ramey said that "our advisors in government, science, and industry have been led to the conclusion that the program must be carried at an intensive level through fiscal year 1969." (4) He then covered the present program in detail.

*Throughout this review, numbers in parentheses after quotations will refer to pages in the published text of the hearings.

†AEC.

The first pilot-plant facility, the MPDI at Gloucester, Mass., began operating in March 1965. Four irradiators have already been built and installed, three others have received AEC support through provision of radiation sources, and six other irradiators are in various stages of construction.

Of 21 petitions submitted to the FDA for approval of irradiation of specific food items, 13 have been approved and the rest are pending. Current results indicate that irradiation of a variety of fish, clams, shrimp, and crabs can extend refrigerated shelf life by 1 week to several weeks and still provide a product completely acceptable to the consumer. Of the fruits tested, bananas and strawberries show the best results to date. Low radiation doses effectively disinfest grains, processed cereal products, and tropical fruits such as papayas, mangoes, and pineapples. The economics of potato-sprouting inhibition are still in doubt, pending larger scale storage tests, although there are no doubts about the effectiveness of the method.

Two problems have required considerable attention. The first of these is the potential hazard of the microorganism *Clostridium botulinum* type E. The organism has caused deaths in cases where seafood was improperly treated with standard food-preservation techniques. The second problem is that studies in fruit preservation do not lend themselves to extrapolation. What is true for one species may be quite the reverse for another very similar species or even a different variety of the same species.

Several steps have been taken to accelerate industry participation in the program. A dozen companies have formally expressed a desire to cooperate with the government in shipping, storage, and laboratory evaluation tests of radiation-processed seafood. Discussions with the Army and industry have also been in progress over a joint large-scale meat-irradiation facility.

In late 1964 the AEC requested the Advisory Committee of the American Institute of Biological Sciences to evaluate the current status and future needs of the program. The committee concluded that the program should be extended at least through 1971; the AEC concurred. The extension would encompass product development and the necessary comprehensive wholesomeness tests for the FDA. Parallel studies of marketing and economic factors would be carried out in conjunction with the Army and other agencies.

Specific Review of AEC Experiments, FDA Status, and Irradiators

By E. E. Fowler*

Mr. Fowler submitted to the JCAE detailed reports on all the experiments so far, including 17 contract summaries (pp. 15-20), product status (pp. 21-22), FDA petitions (p. 23), and data on the current development of irradiator design (pp. 24-29). His testimony indicated that strawberries are the single most promising fruit for application of irradiation processing. Results on peaches vary widely, even with crop years. With doses above 0.2 Mrad, oranges can develop off-flavors, but these disappear in storage. Bartlett pears show ripening inhibition at higher doses than other fruits, probably because ozone is produced in the irradiated fruit. The inhibition effect on tomatoes depends very strongly on their degree of ripeness. Fresh figs respond favorably to radiation treatment; dried figs and prunes also show promise. Papayas and pineapples can be rid of fruit fly infestation at low doses, and mangoes are disinfested at slightly higher doses—all of which could mean a developing export market for Hawaii. Asparagus deformation after harvest is apparently inhibited by irradiation, and bananas show a marked inhibition of ripening which is easily counteracted by gas treatment.

The shelf life of haddock and soft-shell clam meats can be doubled with no effect on quality. Army troops at Fort Lee, Va., could not tell the difference between the 30-day-old irradiated food and top-quality fresh-frozen haddock. Pacific crabmeat and flounder and petrale sole were very responsive to the process. Irradiation of shrimp eliminated "black spot," but a possible problem is the condition of the product when it is received in port. Cod and ocean perch responded favorably to irradiation, but, at doses above 0.2 Mrad, oysters produced a yellow exudate that altered their appearance most unfavorably. Freshwater fish have also been tested favorably. Fowler pointed out that pollution of our coastal waters is making it increasingly difficult to meet microbial standards with usual methods of treatment. This

*Acting Director, Division of Isotopes Development, AEC. (Appointment confirmed on Dec. 13, 1965.)

makes treatment by radiation even more attractive.

The development of suitable irradiators is considered vitally important to the program. Four pool type research irradiators have been installed at different universities, and three others are planned. Two shipboard irradiators have been built, and a third unit will be built for land use. The Marine Products Development Irradiator (MPDI) became operational in March 1965. A Grain Products Irradiator (GPI) and a Mobile Gamma Irradiator (MGI) were scheduled for operation by the end of 1965. Fowler said that commercialization of irradiation processing of strawberries should be accomplished by the spring of 1967, as a result of application of the MGI in California. Another irradiator of "semiproduction" design is scheduled for Hawaii.

Review of Wholesomeness Work

By Leo A. Whitehair*

Dr. Whitehair submitted a statement on the AEC Division of Biology and Medicine program on wholesomeness and public-health aspects of irradiated foods. Summaries of the 19 contracts now in effect were presented (pp. 31-38).

In the discussion that followed Whitehair's report, Rep. Craig Hosmer asked whether or not industry response so far had been "wildly enthusiastic." Commissioner Ramey replied that, of the 20 companies that have shown interest, 5 are spending more money on research with irradiation than on any other new form of food processing, 10 are spending an equal amount, and 5 are spending barely enough to maintain current awareness of developments.

Hosmer called on Fowler to explain the status of the demonstration programs. Fowler replied that industry participation is highest in such programs as the MPDI. Fruit irradiation is purely in the research category. Meat-irradiation research remains with the Army, although a proposal to establish a joint AEC–Army pilot plant with industry is current.

Ramey was then asked to project AEC fund levels. He said that they should remain the same through 1971 as they are now, roughly $2 million to $2.5 million. This would not include the meat-irradiation pilot plant. Consumer acceptance, as brought out by Hosmer, is considered a problem of industry, although the AEC will cooperate. Hosmer also questioned the applicability of this technology to the food problems of emerging nations and came to the conclusion, "I guess it is safe to say that it is not part of the foreign aid program." (43)

The next problem (one that kept appearing throughout the hearings) was the price of ^{60}Co. John Conway, Executive Director for JCAE, claimed that the U. S. price was double that in Canada. Fowler refuted this, pointing out that for 100 thousand curies or more the prices were the same.

[Enclosure 3, pp. 566-590, showed that USAEC led AECL in price reductions. Cobalt-60 was reduced to $0.50 per curie by USAEC in September 1963, and this reduction was followed by AECL only in March 1965. For both agencies, this applied to orders of over 100 thousand curies and did not include final encapsulation costs.]

Hosmer asked about review practices of the AEC in regard to its contractors' reports and about the availability of the resulting information. He seemed to feel that at some point the AEC would have, if it did not already, "an unmanageable and unusable mass of information in the filing cabinet." (46) George Kavanagh, Assistant General Manager for Research and Development, AEC, in defending AEC practice, pointed out that food irradiation is still a small and concentrated field. Moreover, far larger fields of knowledge such as the space sciences are still "in the early stages" of setting up their information programs for large-scale operations. Hosmer claimed, "It's a rat nest out there in Germantown" (46) as far as getting information goes. [This remark received quite a bit of publicity.[20] Its refutation received none. In Enclosure 5, pp. 591-592, Kavanagh pointed out that the AEC receives monthly, quarterly, and yearly technical progress reports from all contractors. At least 65 different annual reports are distributed to more than 450 agencies, companies, or libraries that have expressed interest in this technology. In addition, several hundred copies are sent to individuals who wish to receive the information. Another 26 annual reports summarize work on wholesomeness.]

Rep. William McCulloch asked for clarification of authority on product safety. Ramey replied that final clearance is entirely in the hands of FDA but that AEC programs continue to investigate this problem.

*Division of Biology and Medicine, AEC.

Sterilization Standards and *Clostridium botulinum*

By Samuel A. Goldblith*

Dr. Goldblith submitted a formal statement and, in addition, gave his testimony informally—a practice followed by nearly all witnesses. His two major points concerned the 12-D concept and the superiority of radiation treatment to suppress *C. botulinum* type E. Suppression of *Salmonella* was also mentioned. The efficacy of radiation preservation as an international force to improve standards of living was emphasized.

The 12-D concept comes from work by Esty and Meyer[23] on thermal death-time curves for *C. botulinum*. They loaded test tubes with 6×10^{10} spores each and determined the resistance of the spores to heat. The time necessary at 250°F to reduce the total number of spores to less than one per tube, or a reduction by 12 orders of magnitude, was determined.

Goldblith claimed that (1) if such a concept were actually used in the canning industry, most food would be too overcooked to eat; (2) the concept of 12-D is unnecessary because it would be most unusual if 10^{12} spores of *C. botulinum* were ever present in food. Moreover, if irradiation processing had come along first to establish this standard, heat processing would not be able to make the grade. He then discussed the rumor that destroying normal spoilage flora with substerilizing doses of radiation enables botulinum organisms to grow and flourish. Seven deaths in 1963 were attributed to the presence of *C. botulinum* type E in commercially processed fish. Goldblith pointed out that this organism is not known to appear in fresh fish and that irradiation processing is combined with refrigeration to keep fish in the fresh state. He reported that extensive experiments for the FDA in 1962–1963 failed to turn up any evidence of botulinum organisms. And he said, "If one is to state that radiation 'pasteurized' marine products are a public health hazard due to the possible outgrowth of botulinum organisms because of the altered microbial flora, then the same analogy used to develop this statement must dictate, even more strongly, that thermally pasteurized milk is even a greater public hazard." (52)

He pointed out that ionizing radiation shows great promise for destroying *Salmonella* in poultry, eggs, and other poultry products; in treatment of fish, bone, and blood meal for animals; and in coconuts, spices, condiments, and meats for humans. Irradiation is, he feels, the only means of solving the problem of salmonellosis.

The value of radiation treatment of foods as an international force for cooperation in raising living standards is indescribable. Dr. Goldblith mentioned eight laboratories and two associations in Japan, nine laboratories in Europe, and one each in Israel and Canada—all studying various aspects of food irradiation.

In the discussion period that followed his testimony, Goldblith stressed that he was not proposing the 12-D concept. The concept was used when the first petition to FDA to approve irradiated bacon was submitted. No one sets such a standard at present, especially for canned meats, which usually use only a 5- to 7-D level of treatment.

Rep. William Bates noted that money was authorized by Congress over a year ago for consumer acceptance studies on both irradiated wood plastics and food; however, although the wood–plastics study is well under way, the Bureau of the Budget has not yet released the money for the food study. Kavanagh said that the food study involved a questionnaire that the Bureau of the Budget thought to be too early. The money should be released after further clearances are given by FDA. Marketing work is being carried out by Western Nuclear Co. on potatoes, and the USDA is starting studies on marketing of radiation disinfestation of wheat.

Toxicological Safety of Radiation Preservation

By Herman F. Kraybill†

Dr. Kraybill spoke on wholesomeness and toxicological safety of low-dose radiation preservation. He noted that no other food preservation process has ever undergone testing of such severity. The possible dangers that are being considered in evaluation tests are induced radioactivity, breakdown products in the food, radiation-degradation products in food

*Department of Nutrition and Food Science, Massachusetts Institute of Technology.

†American Institute of Biological Sciences (AIBS)–AEC Advisory Committee.

additives, radiation-induced products in packaging material, nutrient alteration, toxin formation, and production of carcinogenic mutagens from long use of irradiated foods. So far, none of these adverse pathophysiological aberrations have been observed in either radiation-pasteurized or radiation-sterilized food, but further experimentation is in progress.

Conway mentioned research by Monsen[24] who found an auricular rupture in mice used in feeding tests, but Kraybill pointed out that this was a genetic artifact of the strain of mice used. Further tests have not shown such an effect in other experimental animals. Rep. Thomas Morris was careful to check on the manner in which the animals were used. When Kraybill assured him that treatment was humane, he said, "We have all kinds of intellectual organizations in this country, who are making issue of everything that is brought up. I just want to make sure that the record is clear on that point." (74)

Review of Marine-Products Irradiation Program

By Joseph W. Slavin*

Mr. Slavin testified on radiation pasteurization of fishery products. He mentioned that this is one of the few programs with which he has been associated where there was such excellent communication between researchers at different institutes and in different disciplines. He said that irradiation can double or triple the refrigerated shelf life of 16 species of fish and shellfish. Species respond somewhat differently, but results are uniformly encouraging. At the low doses used, bacteria eventually grow back to spoil the product, and he emphasized that this is very important because it enables the consumer to associate irradiated fish with fresh fish, since both spoil.

The quality of the fish used in the experiments was found to be a significant factor, and this has led to the concept of the shipboard irradiator to get the fish irradiated at maximum freshness. The MPDI is presently operational. A cooperative program with industry is the next stage, with industry evaluating shipping and storage procedures; consumer acceptance studies come afterward. Slavin urged that consideration be given to a fleet of fishing vessels that would use nuclear energy not only for irradiation of the catch but also for propulsion.

Slavin concluded that the research is valuable because it strengthens the fisheries industry economically, provides the consumer with high-quality food, enables the armed forces to be better fed, and makes better use of our ocean resources possible. Slavin showed the committee some irradiated-cod fillets 13 days old that were as good as fresh fish kept 1 or 2 days in the refrigerator.

Under questioning by Chairman Holifield, Slavin estimated that application of the process would double the present seafood market. He also brought out that the current industry-cooperation project would be carried out in several phases. Industry would provide fish to be irradiated and would evaluate it in their own laboratories and plants and rate it for quality changes after storage. The final step would be evaluating shipping procedures.

Slavin also mentioned combining heat, radiation, and chemicals in one treatment to really revolutionize the food-processing industry. When questioned closely, he admitted that little work has been done in this area; however, the prospects are very good. Senator Bourke Hickenlooper asked him to compare radiation pasteurization and freeze-drying. Slavin said that, in general, freeze-drying is more expensive but that, for some specialty items, it was probably a bit better.

The testimony for the balance of the first afternoon concerned the Army meat-sterilization program and was presented by several researchers led by Willis M. Hawkins, Assistant Secretary of the Army.

Review of Army Meat-Sterilization Radiation Program

By Edward S. Josephson†

Dr. Josephson stated that the military objectives remain the same, namely, to use irradiated food in garrison rations where appropriate, to eliminate costly refrigeration requirements, and to develop lightweight flexible containers for the irradiated food. He in-

*Director, Technological Laboratory, Bureau of Commercial Fisheries.

†Associate Director for Food Irradiation, Food Division, U. S. Army Natick Laboratories.

serted into the record a tabular summary of food items and food-packaging materials approved by the FDA as of April 1965 (Table III-2).

The Army Medical Service concluded studies on wholesomeness of 21 foods treated successfully with radiation. Sterilized foods were ground beef, pork loin, bacon, shrimp, codfish, chicken, tuna, beef stew, chicken stew, carrots, coleslaw, corn, green beans, sweet potatoes, fruit compote, evaporated milk, peaches, and pineapple jam. Flour, potatoes, and oranges were treated with pasteurizing doses of radiation. Dr. Josephson further stated that in the future there will be only minimal need for additional wholesomeness tests.

Other achievements of the Army program during the past 2 years are establishment of linear accelerators as an alternative to ^{60}Co sources, isolation and identification of the three compounds that cause the odd flavor of irradiated beef, selection of six classes of plastic packaging material with improved radiation resistance, determination of radiation resistance of many *C. botulinum* strains, improvement of beef palatability by irradiating at low temperatures and by packing in vacuo at temperatures of from 38 to 65°C, development of irradiation capability at controlled temperatures from −196 to 90°C, and improvement in uniformity of radiation dose.

Senator Hickenlooper asked for clarification of "radiation resistance of many strains of *C. botulinum*." Josephson said they had determined that the most resistant strains of *C. botulinum* were killed when irradiated in buffer solutions at meat sterilization doses. They logically assumed that the less resistant strains were killed too.

Josephson introduced into the record what he termed "the most up-to-date scientific review of the food irradiation field." (93) This was the proceedings[25] of a conference in Boston sponsored by AEC, U. S. Army Natick Laboratories, and the Advisory Board on Military Personnel Supplies of the National Academy of Sciences. Thirty-seven papers were presented, with good representation from foreign countries and international agencies. Next he reviewed in detail the financial structure of the Army food-preservation program of the last 6 years and submitted a summary of contracts and project orders currently in effect (pp.107-111).

The Army's primary research goal is radiation sterilization of seven meat items in flexible lightweight packaging, and Josephson indicated that the future of the process depends on the capability of the food industry to produce the required commodities. For this reason, invitations to bid on a contract to provide 30 thousand pounds of irradiated bacon were issued to industry in early May. Rep. Price elicited the statement from Josephson that there were only two "highly hedged" responses.

The Army and AEC are currently working on a pilot-plant proposal for processing meats and poultry. Although industrial participation is the key to the future, the Army will continue research in four specific areas: (1) collation of data for petitions to FDA on sterilized meats, poultry, and fish; (2) development of flexible lightweight packaging; (3) improvement of troop acceptability; and (4) study of fundamental problems such as delineation of microbiological and radiation dosimetry standards; improvement of flavor, color, and texture; and achievement of radiation sterilization of completely raw meats, poultry, and fish. Josephson stated that they are using the 12-D concept now, but a modified standard would make radiation sterilization more attractive commercially and the irradiated product more acceptable.

He presented the Army's plans through FY 1971. Current estimates indicate that by the end of FY 1970 the technical feasibility of the process will have been fully demonstrated and a complete series of petitions to FDA will be in progress. During the exchange that followed, Rep. Price congratulated the Army for putting their funds on a 5-year basis; he wondered why AEC did this in everything except this particular program.

Josephson's testimony concluded with his estimate that development of irradiated combat rations, as differentiated from garrison rations, would commence in 1968 and thereafter "coincide with the production capabilities of an industry-sponsored pilot plant and with completion of the series of FDA petitions." (101)

Chairman Holifield questioned the extent of Army involvement in the food-irradiation program. Hawkins replied that the Army is wholly dependent on industry for its food. Therefore the Army must show industry that the process is not only feasible but also profitable. However, efforts should concentrate on "things that

Table III-2 ITEMS FROM THE FOOD-IRRADIATION PROGRAM APPROVED BY THE FOOD AND DRUG ADMINISTRATION (FDA) AS OF APRIL 1965*

Product and purpose of irradiation	Radiation source	Dose (permissible range), Mrads	Petitioner	Date of FDA approval	FAP No.	21 CFR reference
Bacon: purpose, sterilization	^{60}Co	4.5 to 5.6	Army	Feb. 8, 1963	890	121.3002
	Electron beam (5 Mev)	4.5 to 5.6	General Electric Company†	Aug. 23, 1963	1139	121.3004
	^{137}Cs	4.5 to 5.6	AEC†	Jan. 30, 1964	1226	121.3002
	X rays from electron beam (5 Mev)	4.5 to 5.6	Radiation Dynamics, Inc.†	Dec. 15, 1964	4M 1433	121.3005
	Electron beam (10 Mev)	4.5 to 5.6	Army	Apr. 15, 1965	1205	121.3004
Wheat and wheat products: purpose, insect deinfestation	^{60}Co	0.02 to 0.05	Private citizens†,‡	Aug. 21, 1963	941	121.3003
	^{137}Cs	0.02 to 0.05	AEC†	Oct. 2, 1964	5M 1509	121.3003
White potatoes: purpose, sprout inhibition	^{60}Co	0.005 to 0.010	Army	June 30, 1964	1132	121.3003
	^{137}Cs	0.005 to 0.010	AEC†	Oct. 2, 1964	5M 1509	121.3003
Packaging materials§: purpose, food contactants for use in radiation preservation of prepackaged foods	Gamma emitters	1.0 maximum	AEC	Aug. 10, 1964	1297	121.2543
Vegetable parchment paper: purpose, food contactant for use in radiation preservation of prepackaged foods	^{60}Co	6.0 maximum	Army	Mar. 8, 1965	5M 1622	121.2543
	^{137}Cs	6.0 maximum	Army	Mar. 8, 1965	5M 1622	121.3002
	X rays from electron beam (5 Mev)	6.0 maximum	Army	Mar. 8, 1965	5M 1622	121.3005

Note: Mev = million electron volts; FAP No. = FDA food additive petition number; CFR = *Code of Federal Regulations*.

*Ref. 17, page 103.

†The Army contributed the major portion of the data submitted by the petitioner.

‡L. E. Brownell, University of Michigan; T. Horne, Curtiss-Wright Corp.; W. J. Kretlow, University of Michigan.

§Nine classes of materials: nitrocellulose-coated cellophanes; glassine papers; wax-covered paperboards; polypropylene films; ethylene–alkene-1 copolymer films; polyethylene films; polystyrene films; rubber hydrochloride films; vinylidene chloride–vinyl chloride copolymer films.

the Army needs—the things for garrison feeding and the things for field ration feeding." (115)

The next question from the JCAE concerned the degree of coordination between the Army and the AEC; Hawkins termed it "absolute." When Chairman Holifield asked if they got all the money asked for, Josephson replied he was "so happy that I can hardly believe it." However, Hawkins pointedly said, "He is the only one in the Army that is, apparently." (116) When Holifield persisted in this line of questioning, Hawkins commented, "My budget guardian angel whispers in my ear we have not seen the appropriations yet." (116) There are always differences between the budget the President submits and the one Congress approves, of course. Rep. Price brought out in questioning that despite FDA approval, the Army is still using irradiated bacon only experimentally. In response to another question, Col. E. A. Rosenberger of the Army Surgeon General's office stated, "It is the policy of the Army Medical Service to await approval from FDA before recommending inserting these items into Army rations." (119)

Evaluation of Food-Irradiation Program

By Ralph G. H. Siu*

Dr. Siu made four points. (1) The state of the art of irradiated food is one of confidence and maturity. (2) The elimination of the "so-called wet dog flavor of irradiated beef" is an outstanding example of the excellent quality of work done by laboratories and workers throughout the United States. (3) The transition from research to commercialization is very encouraging. (4) The JCAE deserves praise for their continued support of the program.

While the JCAE could not get Siu to be more specific, he did say that "the next step by somebody somewhere will be pilot plant experience." (121) Rep. Price pointed out that to encourage industry to sink funds into such a project, bigger orders than 30 thousand pounds of irradiated bacon would have to be issued. That would be, in his estimate, only $23,000. "They would have to have more of an incentive than that." (121)

*Scientific Director, Research Division, Army Materiel Command.

Review of Fruit-Irradiation Program

By Edward C. Maxie†

Dr. Maxie brought the JCAE up to date on radiation preservation of fruit. He pointed out that the irradiation of fresh fruits and vegetables involves dealing with living systems. "Whether irradiation will work depends on the relative sensitivity of the host and the parasite to radiation. If a fruit is tolerant to radiation and the pathogen sensitive, we may be able to use irradiation effectively." (122) He complimented the AEC divisions of Isotopes Development and Biology and Medicine on the progress made under their programs. He termed the research irradiators and accessory facilities "superb." He outlined several problem areas such as "the almost comical assumption that freedom from rot is synonymous with shelf life." (123) He emphasized, "With the exception of a few papers by USDA scientists and foreign workers, scientific evaluation of the irradiation of fruits and vegetables dates from the entry of the Commission into the field." (123)

Maxie illustrated part of the difficulty: lemons subjected to 0.2 Mrad decay faster, while oranges given the same dose can be stored nearly four times as long as is normal in the trade. Freestone peaches often are severely damaged by gamma radiation even though nectarines, a mutation from the same species, show much less adverse response. Ripening of Bartlett pears is irreversibly inhibited, but stimulated in peaches and nectarines by the same dose. Differences in growth cause different responses to irradiation. "Partially ripened Bartlett pears show little or no response to the same doses that irreversibly inhibit the ripening of completely green fruit." (124) Since few rot organisms can penetrate the protective skins of fruit, the limiting factor will probably be radiation injury to the skin rather than insensitivity of the fungi.

Strawberries can be held to 5% infection during intracontinental transportation. This will result in $5 million gain to the industry. Gros Michel bananas can be easily inhibited from ripening. This is important to some emerging nations, because this means they can ship bananas without having to refrigerate them. Radiation can disinfest the papaya and the mango with no effect on fruit quality; this should

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greatly stimulate the fruit industry in Hawaii. Fresh figs have reached the commercial-test-shipment phase, and dried figs are more tender after irradiation. Recent experiments at two universities have shown that irradiated fresh prunes are more tender and cook quicker. Dried prunes kept 6 weeks at room temperature after irradiation show much the same response as fresh prunes. Oranges respond favorably to low doses. Irradiation of tomatoes has shown enough promise to warrant semi-commercial test shipments in 1966. Recent USDA tests indicate that low doses of gamma rays can retard undesirable morphological changes in mushrooms without seriously affecting quality.

Maxie said, "An extensive search of the literature revealed no information to indicate that wholesomeness will be a problem. Texture of the fruit in almost every case will limit the application of irradiation before any changes in vitamins or other nutritional characteristics of the fruit appear." (126) He concluded his testimony with an appraisal of machine irradiators. They will be valuable for fruit when radiation to skin is beneficial but internal radiation is harmful.

Rep. Price asked why a 1963 petition to FDA for clearance of oranges had not been approved. Maxie said that lemons were included originally and that apparently one researcher had reported fruit irradiated in boxes rotted more rapidly than fruit irradiated individually. These difficulties have been cleared up and the petition resubmitted. Price queried him further on machine vs. isotope irradiation. Maxie claimed that he was not competent to make such a judgment, but he indicated that machine irradiation requires tumbling the fruit to get uniform distribution of dose.

The second day of the hearings opened with remarks by Rep. Thomas G. Morris, presiding. He stressed the fact that although the Army and the AEC seemed to be working closely together, Army had funding plans for 5 years while the AEC was on a year-to-year basis; he termed this difficult to understand.

Radiation Disinfestation of Fruit

By Edgar A. Taylor*

Mr. Taylor's testimony concerned using radiation to prevent insect pests foreign to U. S. mainland agriculture from reaching this continent. Hawaii presents a serious problem since it is already infested with three different fruit flies—the melon fly, the Mediterranean fruit fly, and the oriental fruit fly. "A quarantine treatment that can be used on a wide range of subtropical fruits and vegetables, with effectiveness that will not be influenced significantly by the composition or texture of the produce, or type of packaging, will benefit the entire economy of Hawaii." (131) One estimate of the mango market alone in the United States is $13 million per year.

The treatment need only be sufficient to eliminate the reproductive potential of the tropical fruit flies; complete kill of the flies is not necessary. Extensive tests in Hawaii have shown that exposure doses of 200 thousand r will prevent pupation of fruit-fly larvae, and doses of only 15 to 20 thousand r delivered to eggs and larvae will cause pupae to die. Control of irradiation environment was shown to be significant, too.

A research irradiator has been installed at the University of Hawaii and pilot-plant research is under way. "Our hope is that approval for a papaya radiation treatment, our first goal, may be possible within the next 2 or 3 years." (133)

In answer to a question by Rep. Morris, Taylor pointed out that these disinfestation doses are extremely low. Tripling the dosage would have little if any effect on the fruit itself. When Morris pressed this point, Taylor asked Maxie to support him, and Maxie said he was in full agreement. Morris asked for details of the program. Taylor said it started about 1949 but was diverted because the fruit-fly program was considered very important, and at that time radiation treatment was not thought to be as effective as fumigation. In any case, only one man was working on irradiating fruit. About 1955 work was resumed with an irradiator that could only hold two or three mangoes at a time. The radiation sterilization of insects is known to be a very effective procedure; the screwworm has been almost eradicated by such a technique.

In answer to a question by Rep. Morris about future plans and his opening remarks, Taylor replied that the USDA certainly intends to push this program. "The Department is so tired of these fumigation treatments and revisions of schedule that if we could hit on something like this it would be delighted." (136)

*Assistant Chief, Fruit and Vegetable Insects Research Branch, Department of Agriculture.

Statement of FDA Responsibility

By Robert S. Roe*

Mr. Roe summarized the responsibilities of the FDA in the food-irradiation program. The FDA does not engage in the development of processes; its participation in the program is a result of the food-additive amendment of September 1958 to the Federal Food, Drug, and Cosmetic Act. The amendment "includes regulation of any source of radiation intended for treatment of foods and deems intentionally irradiated foods to be adulterated unless such use of radiation has been cleared for safety by regulation. . . ." (137) As Roe put it, "Specifically, in the case of radiation treatment of food, we need to know whether the treatment results in any induced radioactivity in the food, whether it produces toxic substances in the food, or significant nutritive changes." (138)

The first petition to FDA on the use of radiation concerned the application of inspection and control equipment that used isotopes. The bacon clearance started with a petition by the Army, July 11, 1962. Data showed wholesomeness, destruction of massive doses of injected *C. botulinum*, no induced radioactivity, and no carcinogenic effect. The authorization, effective February 1963, has since been amended to include ^{137}Cs in addition to the original ^{60}Co. The petition for insect disinfestation of wheat was submitted September 1962, with the same kind of data as in the bacon petition. Packaging materials have been evaluated, and current petitions before the FDA include treatment of oranges to inhibit surface organisms (see previous testimony of Maxie), disinfestation of wheat with an electron beam, increase of dose used by the Army to inhibit sprouting, and increase in maximum dose for some packaging materials used by the Army.

Rep. Hosmer inquired about the "actual effect of the FDA's ruling both as a legal matter and as a practical matter." (141) Roe said that issuance of a clearance meant that "a product prepared by a process under the limitations set out in the regulations would be safe and that a manufacturer who wanted to undertake that process properly can do so provided he does follow the regulations." (141) In answer to Rep. Morris, Roe pointed out that once food is cleared for a given process, this does not mean that the procedure is not reviewed periodically. FDA officials can and do simply pick cans off the shelf and test them. Labeling requirements were questioned by Morris, too. Roe pointed out that the matter is under consideration. As he put it, "We don't want to devise a label that will simply result as a scare." (145)

*Director, Bureau of Scientific Standards and Evaluation, Food and Drug Administration.

Review of Department of Commerce Activities

By Anthony A. Bertsch†

Mr. Bertsch said that the purpose of his committee is to assist in the early commercialization of radiation-preserved foods. The functions of the committee are to provide motivation and broad guidance for the program, to achieve the highest level of participation by government agencies and industry, and to effect transition of program responsibility from the Army to other agencies and industry as fast as possible. The 10 member agencies of the committee are: National Aeronautics and Space Administration (NASA); Departments of State, Army, Interior, Agriculture, Commerce, and Health, Education, and Welfare; AEC; Small Business Administration; and Agency for International Development (AID). The consensus of the committee is "that even though many products appear to have excellent prospects, commercial feasibility has yet to be established." (147) Plans are being made for a conference in the fall of 1966.

Bertsch also cleared up some of the confusion surrounding a proposed customer survey about irradiated fish (pp. 63-64). He said that the AEC and the Bureau of Commercial Fisheries had withdrawn their request since the Bureau of the Budget felt that (1) "the proposed survey is not needed in order effectively to inform the public with respect to irradiated fish," and (2) "such a survey would be premature since the FDA has not yet approved commercial distribution of the product nor has it received a petition to do so." (148)

The State Department is interested in food irradiation because of the international potential of the process, AID because of its overseas aid to emerging nations, and the Small Business Administration because a share of any commercial market that develops out of irradiation

†Chairman, Interdepartmental Committee for Radiation Preservation of Food, Department of Commerce.

processing of food should go to small business. NASA believes that there is application of the process for food supplies on long-range space flights.

Bertsch introduced into the record copies of two recent reports by USDA: "Economic Feasibility of Radiation-Pasteurizing: Fresh Strawberries, Peaches, Tomatoes, Grapes, Oranges, and Grapefruit" (August 1963) and "Radiation Pasteurizing Fresh Strawberries and Other Fresh Fruits and Vegetables: Estimates of Cost and Benefits" (March 1965) (pp. 255-284 and 467-494, respectively). The first reports results of a survey of produce handlers. "A high percentage of them thought successful market introduction of radiation pasteurized fruits and vegetables would depend on an effective educational or promotional program to acquaint consumers with the process, its value, and its safety." (149) The later report shows that "a profit of 1.2 cents per pound could be realized from controlling strawberry spoilage by irradiation." (149) He also mentioned the new grain irradiator being built at Savannah, Ga., and the program to control *Salmonella* in foodstuffs by radiation pasteurization.

Department of Commerce activities are divided between the National Bureau of Standards' Radiation Physics Division and the Food Irradiation Unit of the Business and Defense Services Administration (BDSA). The former serves in an advisory capacity, the latter as the focal point of government–business relations. The first report[26] by BDSA's Food Irradiation Unit, dated January 1965 and published in June, explores the status of 28 food products and 6 irradiation processes. "Seventeen of these products are found to have good or excellent prospects in either domestic or international markets." (151)

Rep. McCulloch was interested in how small businesses would fare in the irradiation industry. Bertsch seemed to feel that they would benefit from the elimination of spoilage. Equipment is expensive for commercialization of irradiation processing, although cooperatives could probably handle this specific difficulty. Conway pointed out that the Interdepartmental Committee was responsible for keeping the Army in the irradiation field back in 1956, and it has taken an active role in the programs ever since.

Results of AIBS Committee Review of AEC Program

By Bernard S. Schweigert*

Dr. Schweigert's committee, after extensively reviewing the overall program, concluded that the radiation-preservation program of the AEC needs to be continued beyond FY 1968. Additional time is needed to resolve three problems: the possible public health significance of *C. botulinum* type E in marine products; the meaning of physiological maturity of fruits after irradiation; and the lack of pilot-plant-size irradiators. All these have delayed the passage of petitions through FDA.

He said that optimism and promise are rightly in evidence. He also said that the coordinated Army–AEC program should have at least 5 additional years of committed support.

Rep. Hosmer tried to clear up the problem of AEC participation in an industrial process, and Schweigert stated that the AEC should stay in charge at least through the FDA approval phase. Beyond that, industry should be dominant.

Review of Radioisotopes Used in Food Irradiation

By Bernard Manowitz†

Dr. Manowitz discussed radioisotopes, mainly ^{60}Co and ^{137}Cs, used in the food-irradiation program. Uniformity of treatment is one of the most important design criteria in food irradiation, and it is more critical for food sterilization than for food pasteurization. "One would like to be able to predict the dose at any point in a finite target with an accuracy of ±5%," he said (161). Analytical methods for simple geometries have been developed at Brookhaven and distributed with computer codes to several hundred architect–engineers and designers in this country.

Cobalt-60 sources are made in England, Canada, and the United States. The U. S. standard is the BNL source, which is a strip of cobalt metal about ¾ in. wide, 1 ft long, and 60 mils thick, doubly encapsulated in thin stainless-steel jackets. The cobalt is encapsulated, ir-

*Chairman, AEC Advisory Committee for Radiation Preservation of Foods.

†Department of Nuclear Engineering, Brookhaven National Laboratory.

radiated at Savannah River, and encapsulated again. Several commercial companies purchase large amounts of ^{60}Co from Savannah River and act as brokers for such sources. "Of the eight research irradiators now in use around the country all use the Brookhaven design but only the first was actually fabricated at Brookhaven. All the rest were fabricated commercially." (165)

Manowitz illustrated the fact that higher specific activity is more economical. He postulated a strawberry irradiator designed to handle about 10 thousand lb/hr, requiring 670 thousand curies of ^{60}Co. At a specific activity of 2 curies/g, the irradiator would require 4000 BNL strips costing $100,000 to fabricate; for 20 curies/g specific activity, 400 strips at $10,000; and for 200 curies/g specific activity, 40 strips at $1600. The additional costs of irradiation and secondary encapsulation are higher for low-activity material because of the larger number of strips to be handled and the problems associated with loading the reactor. With a high-specific-activity source the strawberries stay in the irradiator a shorter time. Moreover, the irradiator itself could be smaller; the high-specific-activity source would be 2.4 ft long and the low-specific-activity source 240 ft long. Cost of irradiator shielding is also higher for the low-activity source, again because of the greatly increased volume. High specific activity does have problems of heat generation and dissipation, and these are currently being studied. The cost of pasteurizing strawberries with current technology is about 2 cents/lb and can probably be reduced to less than 1 cent/lb as irradiator designs improve.

Rep. Hosmer questioned Manowitz about the AEC attitude toward the "continuous rivalry between the isotopic source people and the mechanical source people." (168) Manowitz replied, "We present our point of view, and I expect the accelerator people probably consider us to be biased. After all, we have spent 15 to 20 years in developing isotope irradiators." (169) Kavanagh broke in at this point to say, "Dr. Manowitz is speaking as an employee of one of our labs who works on isotopic sources. I think he is describing his own position very well. The AEC as an agency, however, attempts not to participate as such in this squabble." (169) Hosmer did bring out that AEC supports work with isotope sources and does not support work with accelerators for this purpose.

Hosmer and Morris then pursued the question of Canadian vs. U. S. prices on ^{60}Co. Manowitz testified that while price per curie in large lots is identical, the Canadian price includes secondary encapsulation. The U. S. price does not. Therefore, as he put it, "Talking about a specific batch and putting it in an irradiator one can get it at a cheaper price from the Canadians than one can get from the United States." (170)

[In correspondence (pp. 660-661) with Conway after the hearings, Manowitz made his testimony more explicit. He said, "Restricting this analysis to 100,000 curie lots, for low specific activities (5–10 c/g) encapsulated Canadian ^{60}Co is considerably cheaper than United States'; at 15 c/g (the highest specific activity in the AECL published price list), encapsulated Canadian ^{60}Co is slightly cheaper than United States'; at 30 c/g, encapsulated U. S. ^{60}Co is probably cheaper than Canadian, assuming that since 30 c/g material is not listed by the Canadians it is either not available or more expensive than United States'."

He went on to say, "I hasten to point out that the factors considered here are only partial factors in the real cost of ^{60}Co in a commercial irradiator. Delivery costs, installation costs, replacement costs, and service costs are factors that can be of determining importance."]

Review of Electron Accelerators Used in Food Irradiation

By H. W. Koch*

Dr. Koch discussed food irradiation using intense electron or X-ray beams. The three basic techniques for irradiation processing are treatment with electrons direct from an electron accelerator, treatment with X rays produced by electrons in an X-ray target used with an accelerator, and irradiation by gamma rays from radioisotopes. Even though the government has emphasized isotope sources in research, private industry has used "direct electrons" almost exclusively.

For foods, media must be penetrated and uniformly dosed over a thickness of 4 cm, which

*Chief, Radiation Physics Division, National Bureau of Standards.

is about the limit of electron penetration. Non-food-irradiation processing usually requires less than 4 cm penetration, and electron beams are used because they are more readily available than isotope sources. A 10-kw electron accelerator is practically an "off-the-shelf" item, while the equivalent gamma source of 700 thousand curies of ^{60}Co is a rarity. The purchase of an electron accelerator implies the maintenance of a proprietary production facility, a factor of great interest to industry.

In the power range of interest for food processing (1.5 to 10.0 Mev), there are five types of accelerators: Dynamitron, insulating core transformer, linear accelerator, resonance transformer, and Van de Graaff accelerator. [Koch and Elmer Eisenhower summarized accelerator characteristics in a paper[27] entitled "Electron Accelerators for Food Processing." Results of a survey presented in the paper show that there are more than 500 electron accelerators operating today that are suitable for food irradiation.] There are five advantages of electron processing: (1) The direct deposition of the power in a primary electron beam is highly efficient. (2) The electron accelerator can be turned off. It presents no maintenance problem. (3) Electrons have variability of power. (4) The monodirectional characteristic of the electrons and X rays permits greater flexibility of package design. (5) The ability to program the various beam parameters from one instant to the next means that a greater variety of shapes and densities can be handled automatically.

Koch emphasized that he thinks electron accelerators have been shortchanged in research. Only after complete investigation into their uses can electrons, X rays, and gamma rays be properly evaluated for food processing. Rep. Hosmer was curious about whether government or industry should be responsible for this further research. Koch pointed out that the industries that are prospective users of electron generators want to have more information. "However, they can't get that information simply because the industries that have the information regard their applications as proprietary." (174-175) Hosmer said, "While the spotlight is being occupied by government with a bag of public gold, industry naturally stays on the sidelines and awaits in the wings for any fallout it can get." (175)

Koch also said he thought one reason for the popularity of machines over isotopes was the availability of machines, based on the long lead time needed to get any ^{60}Co. Fowler disagreed, saying that megacurie quantities of ^{60}Co are available now and have been for the past 2 years. He agreed with Koch that proprietary interests play a role in the selection of the machine as a source. Under present rules, if a source is provided with AEC support, the results of the research have to be made public; if a source is bought from a commercial or a government agency, though, the results obtained with that source belong to the researcher.

NUMEC—Current Food-Irradiation Program

By Zalman M. Shapiro*

NUMEC has been involved in the irradiation-processing program for a number of years as an architect–engineer, manufacturer, operator, and commercial investor. Dr. Shapiro said they have built and are delivering two shipboard fish irradiators, and they have been selected to provide architect–engineer services for the Hawaiian fruit irradiator. NUMEC is an equal partner with Israel in Isorad, and 30 thousand curies of ^{60}Co have been shipped to Israel for use in experimental facilities. Shapiro said, "We have initiated contact and correspondence with officials at the federal, state, and local levels with regard to possible support for an irradiation facility." (178) He also mentioned a proposed major experiment in cooperation with Pillsbury Mills, Inc., to be supported by AEC funds. This will be a comparison of storability of potatoes treated with chemicals and with radiation to prepare for successful commercialization of the process. It will be analogous to the "team effort" used by the AEC to demonstrate the feasibility of commercial nuclear power. Shapiro said that funds for a food demonstration program should be specified by AEC and used to gain industrial acceptance, to develop economic data, and to encourage consumer acceptance. One of the steps he suggested to encourage consumer acceptance would be to set up exhibits at fairs and at food and home shows in cooperation with private companies. He termed consumer acceptance "the most signif-

*President, Nuclear Materials & Equipment Corp. (NUMEC).

icant obstacle to the large-scale commercialization of food irradiation." (181) Another of his suggestions was for the AEC to support food-irradiation programs in other countries. Food irradiation could enable emerging nations to take a giant step into the 20th century, perhaps skipping the use of refrigeration. Rep. Hosmer questioned the principle of skipping refrigeration and argued that production was more of a problem than distribution. When he said that 30% of the grain shipped to foreign countries rots on the docks, Shapiro corrected him, pointing out that the grain is consumed by insects and that disinfestation by radiation could correct this.

Specific questioning by Hosmer about the Pillsbury–NUMEC proposal revealed that the total program cost would be $60,000. He said, "Do you mean to tell me that Mr. Pillsbury who gives away $60,000 worth of prizes to housewives in cooking and baking contests can't get $60,000 together to go into this thing?" (185) Shapiro defended his proposal with the argument that the information was to be made generally available. He said, "I think that the food irradiation program is too big to be the sole property of any particular firm. I think it should be the property of the country as a whole." (186) When pressed about the amount of "substantial contribution" Pillsbury and NUMEC would make to the program, he declined to answer since the matter was still in negotiation with the AEC.

Rep. Wayne Aspinall wanted more details on the idea of a pilot-plant system. Would the government run it, with industry able to witness and learn? "I guess that it would depend entirely on the nature of the specific contributions made by the parties involved, as to who really has the principal voice in the operation," said Shapiro (187). Aspinall next asked about cobalt prices and their effect on the food-irradiation program. Shapiro replied, "Generally speaking, as long as the price of cobalt in the United States is higher than the price of cobalt as purchased from Canada or other sources, then I believe it will have an adverse effect on the companies which are designing and constructing irradiators here in the United States." (188) He went on to emphasize that the consumer has to be educated to adjust to this novel method of food processing, especially since "this generation associates radiation with something that is very bad." (188)

Swift & Co.—Current Meat-Irradiation Program

By Walter M. Urbain*

Dr. Urbain expressed views similar to those of other witnesses about the success of the program to date and about the coming commercialization of the process. He specifically pointed out that irradiation facilities have been built and are available, that low-temperature techniques developed by the Army have improved quality considerably, that three foods have been approved by FDA, and that petitions for other foods are in process.

Urbain felt that the next step is "test production" to develop products having commercial characteristics; to work out practical production methods; to develop production-cost data; to determine consumer reaction to quality, utility, the fact of irradiation, and price; to determine marketing requirements; and to develop merchandizing procedures. He stressed the dependence of private industry on the AEC and the Army for the progress made up to this point. "It is my recommendation that to attain it [commercialization], the Army and the AEC be enabled to continue their work as they have proposed at this hearing. Specifically, the work on obtaining the clearance of foods now planned is basic to commercialization of the process." (190)

Under questioning by Rep. Aspinall, Urbain said that Swift & Co. does not have any contractual agreement of any kind with the AEC, although Swift has spent approximately $700,000 of its own in research in the field since 1949. Their present effort is relatively small. He also said that their relations with the Army and AEC are excellent. When asked explicitly about the demonstration program advocated by Shapiro, Urbain said he had not heard of it before. Furthermore, he stated, "I think a necessary first step to that is that a sufficient number of foods have Federal Food and Drug and other agency clearance." (192)

Rep. Hosmer asked about both early and recent research activities of Swift. Urbain explained fully, "Well, we on an experimental basis irradiated various types of meats and determined the effect of the ionizing radiation

*Director, Engineering Research, Swift & Co. (presently at Michigan State University, East Lansing, Michigan).

on the stability of these meats under a variety of conditions. We also identified some of the problem areas such as flavor. We conducted a rather large animal feeding experiment of beef to determine whether irradiated meats were wholesome, and this was one of the first such studies." (192) When it was determined that the process was not commercially feasible and that the Army was starting research on radiation sterilization of meat, Swift cut back its program. It did offer free advice to the Army based on its experience. Later Swift had several no-cost contracts. Current research expenditure is about $10,000 a year according to Urbain, who stressed, "I have been given encouragement to see that Swift keeps abreast of the field and at the right time take action that is needed to use the process." (193) He further stated that in his opinion the process has reached the point where, although industry need not go overboard with investment, there should be plans for increased spending in the near future.

Holly Farms, Inc.—Possibility of Irradiation of Packaged Chicken

By Elmer Stuart Kendrick*

Mr. Kendrick said, "This is a totally integrated poultry meat growing company producing roughly 50 million head of broiler chickens per year and marketing roughly 70 million broilers per year, having its own hatcheries, feed mills, and processing plants." (194-195) The chicken carcass is quickly chilled. All surplus moisture is drained from the carcass in roughly 3 hr. After this, the whole carcass is sealed in a polyethylene wrapper, or it is cut into segmented parts and packaged as a cut-up chicken or as various chicken parts. These packaged products are kept at 28 to 30°F by mechanical refrigeration until sold. The advantages of the process are less handling, governmental supervision of sanitation, completely sealed and uncontaminated product, and transportation in refrigerated sealed units.

The process can be modified to use radiation pasteurization of the product immediately after it is sealed into the polyethylene bag. Kendrick felt that this would kill over 90% of all bacteria in the package, including *Salmonella*. He stressed, as had other industry witnesses, "Private industry cannot spend money researching new ways that food can be irradiated until FDA approves the irradiation of food as being not harmful to the consumer." (195) He suggested that food sterilization by radiation was not likely to be economically sound any time soon.

After his formal statement had been inserted into the record, Kendrick related that Holly Farms had first heard of food irradiation in 1964. He made a trip to the Natick Laboratories at that time, although the Army was concerned only with sterilization, not pasteurization. Since that time his company has kept up with the field, although they have not invested any money in the irradiation process. Like other witnesses from industry, Kendrick said, "I feel that the government should continue to carry the burden of this research work until passage of FDA because no company can do anything to protect trade secrets of this work as such." (196)

Rep. Hosmer asked about the use of funds from the government, if they were available, and about the increased cost of radiation pasteurization in this particular application. Kendrick said that they would rather "go it alone" than have any restrictions. Holly Farms would probably rent an irradiator rather than buy or build one, and the process would increase costs about half a cent a pound. Under questioning from James Graham, Technical Advisor to the JCAE Subcommittee, Kendrick pointed out that a combination of heat and radiation would not work very well either for canned or fresh chicken because of the protein enzymes present.

The Gorton Corporation—Current Fish-Irradiation Program

By Robert T. Love†

Mr. Love represented one of the largest and oldest seafood producers in the United States. He commented, "First, we are committed to the seafood business and intend to remain a leader in it. Second, our 'corporate destiny,' so to speak, has been determined in large measure by new technology in the preservation of seafoods." (199)

The Gorton Corporation has responded to the AEC appeal to members of the seafood industry

*Executive Vice President, Holly Farms Poultry Industries, Inc.

†Director of Marketing, The Gorton Corporation.

by agreeing to undertake a preliminary marketing study this summer, although the company has made no decision to use irradiated seafood or in any way to depart from its present marketing and distribution programs. Love said the marketing study will investigate the fresh-fish-market applications because the fresh-fish market will be developed first if irradiated seafoods fill a need, since over 75% of the domestic catch is sold fresh. Gorton will also investigate quality, using its own research and development group. Consumer panel testing will have to await FDA approval.

Love felt that irradiation processing of fish could benefit foreign countries that have a food problem. Fish is known to have high nutritional value. "Nevertheless, one of the major problems to be overcome will be to establish distribution in hot climates, where there are poor roads, where there are no freezers, and in some cases no refrigeration facilities. In these areas, the lower perishability ratio of irradiated fish may open up possibilities to feed large numbers of people [far] removed from the sea." (201)

After his testimony, he was questioned closely by Rep. Hosmer. His replies revealed that the proposed study was a voluntary response to an AEC appeal to industry and involves no AEC money. Irradiated fish will be shipped to the Midwest, and the study will cost Gorton about $6000. No fish will be consumed; the study will involve only talking to buyers and sellers of fresh fish. If there is an obvious economic opportunity, then Gorton will certainly enter the business of irradiating fish and selling it.

Thomas J. Lipton, Inc.—Irradiation of Dried Soup

By Roy Morse*

In addition to being the largest processor of tea in the United States, Lipton has also been in the dehydrated foods field for over 23 years. One of the disadvantages of dehydrated foods is the amount of time needed for cooking to obtain food sufficiently soft to be palatable. In 1955 work started on a process using ionizing radiation to shorten cooking time for dehydrated foods. United States Patent 3,025,171 was subsequently issued to Charles Schroeder, who was present with Morse at the hearing. A dose of 0.3 to 0.4 Mrad reduces cooking time from 15 to 1 min for the dehydrated vegetables used in soups. Another advantage is the additional microbiological safeguard offered by the irradiation process. So far, Lipton has used several types of machines as well as both ^{60}Co and ^{137}Cs. Morse said, "Availability appears to be no problem for either isotopic or machine sources." (204)

A petition to FDA for clearance of the soup is in process. There is actually a physical change in the product rather than a change to be described by such terms as pasteurization or sterilization, although these are obtained as peripheral advantages. The Armed Forces are interested in dried soups for field rations, and NASA could conceivably use them in the space program.

Morse pointed out that expenditure on research in the food industry is relatively low, about 0.3% of the sales dollar. "I think even if you look at that figure you will find that a great percentage of what the food industry calls research is really quality control and is not really probing for new areas and new products." (206) Some of the reasons for this are that the food industry is a very old industry, "full of a lot of art," and change does not come easily. "It is much easier to change the aerospace industry or the electronics industry than it is the spaghetti industry." (206) One reason industry has moved slowly into the irradiation-processing field is that industry is out to make money; the more money industry has to spend, the more return it wants. Moreover, each participant is especially interested in a unique product or method.

Another, very specific worry of industry is the label. Morse said, "If we have to feature in type the same size as the name of the product the fact that this is irradiated, that may be good or bad, I don't know. My guess will be that it is not going to help us very much." (207) Rep. Hosmer asked why Lipton's policy on government cooperation differed from NUMEC's. Morse again pointed out, "If we are not able to get a patent or if the work we do becomes public information, then we are not very enthusiastic about this kind of work." (209) He was agreeable to the idea of time-sharing a reactor or machine with other companies. However, he stated that Lipton's position is slightly

*Vice President, Research and Development, Thomas J. Lipton, Inc.

different from some other elements of the food industry since Lipton can ship dehydrated vegetables anywhere anytime, quite unlike fresh fish or fresh strawberries.

With the conclusion of the testimony before the JCAE, Rep. Hosmer, as acting head of the committee, summarized the hearings:

> During these hearings, we have heard testimony from the AEC, Army, other Federal agencies involved in this program, and representatives of private industry. The committee appreciates receiving this up-to-date information on the food irradiation program.
>
> During these hearings an interesting proposal has been made concerning the establishment of a cooperative demonstration program. It should be seriously considered by the executive branch both as to its possible merits and possible demerits.
>
> There might well be some diversity of opinion with respect to how close we are to the initiation of the commercialization phase in this country. It appears that we are not there yet, but it is not far off. In view of the long leadtimes required for Food and Drug review of petitions of irradiated foods, it seems to me important that we plan for this step so that we are truly prepared for it when the time comes.
>
> I think it extremely important that this program, which can have such far-reaching effects—not only upon industry but upon the general welfare of the world—continue without any loss of momentum. A program of this nature cannot reach its objectives by the simple expedient of supplying an overabundance of funds. We are not suggesting such an approach. The scientists appear to feel that the present funding levels are adequate and should be continued. They are not asking for any substantial change in funding level for these programs.
>
> However, I believe that for proper planning the AEC should be able to lay out a 5-year program similar to what the Army has done. (210)

(F. E. McKinney)

The Quality and Wholesomeness of Radiation-Pasteurized Marine Products, with Particular Reference to Fish Fillets

By J. W. Slavin,* J. T. R. Nickerson,† L. J. Ronsivalli,‡ S. A. Goldblith,§ and J. J. Licciardello¶

Low-energy gamma irradiation and conventional pasteurization processes act similarly to extend the shelf life of foods under refrigeration. This paper discusses radiation-pasteurized fish in relation to general requirements for pasteurized foods. In addition to reviewing acceptability and wholesomeness research, it discusses spoilage, microbiological contamination, commercial procedures for handling fish, and certain safety aspects of the product. It also compares radiation pasteurization with other pasteurization techniques. A review of the literature on marine-product preservation and recent work on fish fillets indicate that radiation-processed foods are high in quality, acceptability, wholesomeness, and nutritional value. The quality of the foods depends on their biological properties, dose levels, storage temperature and environment, and quality of the starting material. Large-scale acceptability tests have shown that radiopasteurized fillets of haddock, cod, and petrale sole compared very favorably with fresh-frozen controls. Animal and human feeding studies show that a number of types of radiation-processed fish are both nutritious and wholesome. At doses below 0.15 Mrad, microbiological changes in irradiated fish are of the same general character as those occurring in other pasteurization processes; spoilage is just as recognizable in irradiated as in pasteurized products and appears similar. Studies on haddock fillets indicate that fish muscle is a poor substrate for growth of Clostridium botulinum *type E. In experiments conducted thus far, haddock fillets, inoculated with excessively higher numbers of botulinum spores than those found to occur normally, underwent organoleptic spoilage. Other safeguards are common to food-pasteurization processes, including well-established commercial practices for storing and shipping fish at ice temperatures and cooking of the finished product. The process appears to be economical and to offer the consumer a better product.*

The general belief that ionizing radiation might be useful in preservation or substerilization processes dates back some 15 years, although it was early recognized that radiation pasteurization of food might be possible.[28a] One of the more obvious alternatives to the use of sterilizing doses of radiation and attendant disadvantages was the use of lower dosages (under 1 Mrad) to destroy the bulk of the nonspore-forming organisms, leaving only the spores. The product would then be stored at refrigeration temperatures above freezing.

In June 1951 Proctor et al.[28b] began work under a U. S. Navy contract on the use of substerilizing doses of electron radiation for the preservation of beef slices. Licciardello, Nickerson, and Goldblith[29] have kept some meat products under refrigerated storage for 12

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years. Proctor and coworkers,[30] in a feasibility study for the AEC, reported that the preservation of certain marine products offered particular promise, and these and other workers[31] outlined a group of projects aimed at studying aspects of radiation pasteurization at various laboratories.

In these studies, certain aspects pertaining to the wholesomeness of irradiated marine products have been investigated: (1) the effect on the nutritive value of the foodstuff, (2) the question of whether or not any toxic substances are induced in the foods by treatment with radiation, and (3) the microbiological aspects.

Satisfactory resolution of the above criteria is considered sufficient to the requirements of the Food Additives Amendment of 1958 in terms of wholesomeness of irradiated foodstuffs.* Much effort, money, and time have gone into this phase of the irradiation preservation of foods program.

The radiation method has been scrutinized by a large number of scientists, and all the data today indicate that the products are safe and wholesome if the product is held below 38°F. One may question whether or not the standard methods of thermal processing or smoking of foods could undergo such rigorous testing and pass muster.

With respect to the microbiological aspect of radiation-preserved marine products, some scientists have felt that inasmuch as substerilizing doses will destroy many of the nonspore-forming organisms, there would be a better likelihood of outgrowth of any *C. botulinum* type E spores present with concomitant toxin production. Data obtained thus far with extensive inoculated pack studies indicate that there is no more danger of botulism from irradiated substerilized haddock than from normal, untreated haddock products.

Both scientists and regulatory officials recognize that pasteurization must meet criteria often quite different from those for sterilization. As with other food processes, pasteurization also must fulfil certain basic requirements associated with efficacy, or measured benefit in quality extension, in addition to wholesomeness and nutritive value. Furthermore, since pasteurized products are perishable, they must also meet certain conditions to safeguard the public against any possible danger. Among these are some built-in safety factors: (1) discernible spoilage characteristics to permit the consumer to reject low-quality products, (2) long-established hygienic and refrigeration techniques that preclude mishandling, and (3) cooking the product, which reduces the numbers of microorganisms.

*The matter of induced radioactivity is not considered herein inasmuch as it has been demonstrated that electrons of energy level up to 10 Mev, X rays up to at least 5 Mev, and gamma rays from ^{60}Co and ^{137}Cs may be used with impunity.

Quality and Acceptability

A number of different species of fish and shellfish have been investigated to ascertain organoleptically and chemically their response to radiation pasteurization and storage. Information reported at the Fifth Annual AEC Contractors' Meeting on Radiation Preservation of Food[32] indicates that irradiation at doses less than 0.5 Mrad doubles or triples the shelf life of 16 species of fish and shellfish (Table III-3).

Table III-3 OPTIMAL RADIATION DOSE LEVELS AND SHELF LIFE AT 33°F FOR FISH AND SHELLFISH AEROBICALLY PACKED IN HERMETICALLY SEALED CANS

Seafood	Optimal irradiation dose, Mrad	Shelf life, weeks
Oysters (shucked)	0.20	3 to 4
Shrimp	0.15	4
Smoked chub	0.10	6
Yellow perch fillets	0.30	4
Petrale sole fillets	0.20	2 to 3
Pacific halibut steaks	0.20	2
King crab meat (cooked)	0.20	4 to 6
Dungeness crab meat (cooked)	0.20	3 to 6
English sole fillets	0.20 to 0.30	4 to 5
Soft-shell clam meats	0.45	4
Haddock fillets	0.15 to 0.25	3 to 4
Pollock fillets	0.15	4
Cod fillets	0.15	4 to 5
Ocean perch fillets	0.15 to 0.25	4
Mackerel fillets	0.25	4 to 5
Lobster meat (cooked)	0.15	4

The optimum dose—that which gives greatest shelf life without changing normal characteristics of the product—for most of the species studied varies from 0.15 to 0.30 Mrad. Exceptions are clams and smoked chub, for which the optimum doses are 0.45 and 0.10 Mrad, respectively. But even when these are included, the optimum dose for the seafoods studied falls within a relatively narrow range.

We would like to point out, however, that these were experimental, not commercial, storage

studies. Products were packed in hermetically sealed cans, in air, or under vacuum and were tested by expert sensory panels after irradiation and a defined period of refrigerated storage. It is only recently that any storage and acceptability studies have been carried out on products packed under commercial conditions.

QUALITY STUDIES

The quality and subsequent storage life of irradiated seafoods are influenced by (1) inherent biological makeup of the product, particularly its lipid composition, (2) dose levels, (3) gaseous environment and temperature during treatment and storage, and (4) quality of raw material.

Biochemical Composition. The response of a product to irradiation is significantly influenced by the biological properties of the starting material. Lipids have a pronounced influence on acceptability, as evidenced by the rancidity of the lipid fraction in petrale sole, halibut, and swordfish. However, the fact that some fish, such as mackerel, that are rather high in fat are amenable to irradiation indicates that the composition of the lipids, rather than the amount, may be the critical factor. In addition, since some fish, such as salmon, change color during irradiation, carotenoid pigments might also be affected.

In the species studied, decomposition by endogenous enzymes does not appear to be a significant problem when the refrigerated shelf life is only doubled or tripled. Irradiated clams, which were most affected by endogenous enzymes and became slightly tender after 30 days of storage at 33°F, were still highly acceptable.

Dose Levels. Dose levels affect quality and shelf life of fish. In general, at levels 1.5 to 2 times the optimum dose, objectionable off-odors are readily recognizable when the products are served steamed and less readily recognizable when served deep-fat-fried. At dose levels less than optimum, product shelf life is reduced, but a significant reduction in shelf life occurs only when the dose is about 50% below optimum.

Organoleptic appraisals have been confirmed by bacterial analyses. We have found that at a dose of 0.25 Mrad, 99 to 99.9% of the bacteria, or 2 to 3 log cycles, are destroyed; whereas at a dose of 0.15 Mrad, 90 to 99%, or 1 to 2 log cycles, are destroyed.

Gases and Temperature. Both product quality and shelf life might be significantly altered by gaseous environment and temperature, and several workers have recommended remedies. Dassow and Miyauchi[33] reported that the development of rancidity in petrale sole and similar fish might be minimized by excluding air from the package during irradiation. Their data indicate that this can be done by vacuum packing or tightly packing in low-oxygen-permeable materials with exclusion of as much air as possible. Also, use of FDA-approved antioxidants or of inert gases to replace oxygen in the package should not be overlooked.

Gaseous environment affects shelf life of the irradiated product, much in the same manner as it does the fresh product. If a product is continually scrubbed with air, most spoilage bacteria will multiply faster and the product will spoil quicker than it would with a limited air supply. For example, we found that irradiated fillets air packed in sealed cans kept several days longer than those air packed in gas permeable plastic pouches.

As would be expected, temperature of storage directly affects the shelf life of the irradiated product. Studies by Spencer and Baines[34] suggest that $Q_{10} = 3$, or that for each 10°C (18°F) reduction in temperature between 25°C (77°F) and 0°C (32°F), the shelf life of white fish was tripled. In almost all cases, irradiated products stored at 33°F have been found to keep about twice as long as those stored at 42°F, which is about the same as for nonirradiated seafood.

Quality of Starting Material. As one would expect, the starting quality of the fish to be irradiated has a significant effect on end-product quality and shelf life. Studies conducted at the University of Washington on sole and at the Bureau of Commercial Fisheries (BCF) Technological Laboratory at Gloucester on haddock and cod are in close agreement and indicate that fillets cut from fish stored up to 7 to 9 days on ice are suitable for irradiation. Figure III-8 shows the effect of preirradiation storage time, which is an indication of quality, on the postirradiation shelf life of cod and haddock fillets. Preirradiation age of the fish does not materially affect storage life of the irradiated fillets until after about 4 days on ice, at which time the shelf life of the treated fillets decreases at an increasing rate with an increase in age of the iced fish.

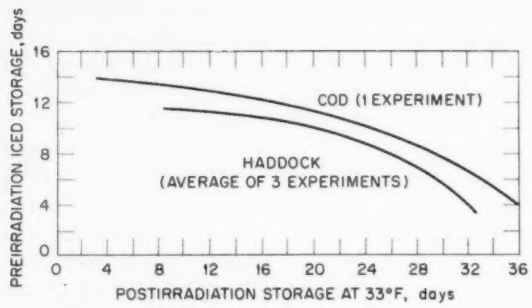

Fig. III-8 Effect of preirradiation age of iced, eviscerated fish on postirradiation shelf life of the fillets stored at 33°F.

ACCEPTABILITY STUDIES

Large-scale acceptability studies have been carried out at Fort Lee, Va., in cooperation with the U. S. Army Natick Laboratories, on radiopasteurized fillets of haddock, cod, and petrale sole. The frozen controls and treated samples were served at regular Army mess to participants who had no knowledge as to which of the samples were irradiated. The acceptability scores shown in Table III-4 show that the irradiated product compared very favorably with fresh-frozen controls.

Wholesomeness and Nutritional Value

When a new preservation process is to be investigated, much attention is always given to the effect of the treatment on the biochemical properties of the food, particularly as they relate to wholesomeness and nutritive value. Improvements in analytical techniques and methodology in recent years have made it possible to scrutinize the safety aspects of new food-preservation methods even more thoroughly than in the past, thus providing the public with greater assurance against the likelihood of possible hazard.

Irradiation is the first food-processing technology that has been subjected to a comprehensive testing and examination of all wholesomeness and nutritional aspects. Long-term feeding studies and nutritional analysis have been carried out on fishery products irradiated at sterilization and pasteurization doses. Results of these investigations all show that the irradiated products are of high nutritional quality and compare favorably with untreated controls.

A complete report of the wholesomeness and nutritive value of irradiated foods was given at recent hearings before the Joint Committee on Atomic Energy.[35]

Table III-4 ACCEPTABILITY TESTS ON RADIOPASTEURIZED SEAFOOD (U. S. ARMY TEST EVALUATION COMMAND, FORT LEE, VA.)

Date of report	Product*	No. of panelists	Dose, Mrad	Storage temp., °F	Storage period, days	Average taste scores†	
						Irradiated	Nonirradiated frozen controls
February 1964	Haddock fillets	314	0.25	33	15	5.76	6.13
					29	5.8	6.17
November 1964	Petrale sole fillets	333	0.20	32–35	16	5.9	6.43
					28	6.52	6.57
May 1965	Haddock fillets	693	0.15	33	7	6.19	5.90
					14	5.99	6.19
September 1965	Cod fillets	588	0.15	33	17	6.59	6.54
					24	6.31	6.23

*All irradiated samples were packed in air in hermetically sealed cans except in the 1965 tests, when products were packed in 30-lb fillet tins under commercial conditions.

†Scores were based on a nine-point hedonic scale where: 9 = like extremely, 8 = like very much, 7 = like moderately, 6 = like slightly, 5 = neither like nor dislike, 4 = dislike slightly, 3 = dislike moderately, 2 = dislike very much, 1 = dislike extremely.

In other evaluations occasioned by ground-breaking and dedication ceremonies for the MPDI, civilian guests voluntarily participated in taste tests on several irradiated products. These people, from all walks of life, judged the irradiated products to be highly acceptable (Table III-5).

WHOLESOMENESS STUDIES

Classical subacute and chronic techniques established by the Office of the Surgeon General, Department of the Army, in conjunction with the Food and Drug Administration, were used as a protocol for tests on the wholesome-

Table III-5 RESULTS OF ACCEPTANCE TESTS BY NONTECHNICAL, CIVILIAN VOLUNTARY GROUPS

Occasion and number of panelists	Product*	Dose, Mrad	Storage temperature, °F	Storage period, days	Average taste scores†
MPDI groundbreaking‡ ceremonies, ~45 panelists	Clam chowder	0.450	33	21	7.4
	Fried clams	0.450	33	21	7.7
	Fried haddock	0.250	33	21	7.6
	Finnan haddie	0.150	33	21	7.4
MPDI dedication, 278 panelists	Clam chowder	0.450	33–35	14	6.4
	Fried haddock	0.250	33–35	14	7.2
	Lobster salad	0.150	33–35	14	7.7

*See first footnote for Table III-4.

†Scores were based on a nine-point hedonic scale where: 9 = like extremely, 8 = like very much, 7 = like moderately, 6 = like slightly, 5 = neither like nor dislike, 4 = dislike slightly, 3 = dislike moderately, 2 = dislike very much, and 1 = dislike extremely.

‡Data were originally obtained on a five-point scale and were converted here to the nine-point scale for reporting purposes.

ness of irradiated fish. In the petition submitted to the FDA for clearance of irradiated marine products,[36] it was pointed out that subacute and chronic animal feeding studies had been successfully concluded on codfish, salmon, haddock, whitefish, and tuna fish irradiated at levels varying from 2.79 to 5.58 Mrads. No abnormal responses or effects were reported in subacute tests with rats fed 35% total solids of irradiated fish over a 2- to 3-month period. In longer-term dog and rat feeding studies lasting at least 2 years, no abnormal responses were noted from animals fed irradiated fish making up, in combination with another irradiated food, 35 to 70% of the total solids of a complete ration. The animals used in these studies were examined periodically for growth, food utilization, and histopathological changes for at least three generations in the rats and one generation in the dogs. Similar findings were reported by Hickman[37] when feeding rats cod fillets irradiated at 0.3 and 0.6 Mrad at dietary levels of 45%.

Wholesomeness data obtained in studies on animals have been confirmed in some limited tests on feeding human volunteers[36] with seafood irradiated at 2.79 and 5.58 Mrads. The effect of irradiated foods, including haddock, on digestive enzyme systems has also been studied in rats.[38] In cases where haddock irradiated at 5.58 Mrads comprised 19.15% of the dry solids in the composite diet fed the animals for their lifetime and during three generations, no differences were noted in assays for liver xanthine oxidase, succinic dehydrogenase, hypotonic endogenous respiration, alkaline phosphatase, and serum alkaline phosphatase. However, a consistent change in cytochrome oxidase activity was noted in the males, but this was attributed to the nutritional factors rather than to low-level toxicity.

As indicated in the marine-products petition submitted to FDA, results of tests using irradiated codfish, salmon, and tuna fish indicated no statistically significant differences in growth, mortality, tumor incidence, and histopathology of control and test animals.

Since irradiated fish treated at levels exceeding 2 Mrads produced no adverse effect on animals or humans fed at high dietary levels for periods up to 2 years, it is logical to conclude that ingestion of fish flesh irradiated at these or at lower levels of pasteurization doses does not result in a public health hazard in terms of inducing formation of toxic substances.

NUTRITIONAL VALUE

In the animal and human feeding studies previously discussed, protein utilization of products irradiated at levels exceeding 2 Mrads was the same as with the nonirradiated controls. Since animals receiving the irradiated fish compared similarly in growth and reproductive characteristics for three generations with those that received the nonirradiated fish, it appears that the nutritional quality of the protein was not affected by irradiation even at these high levels. If high radiation levels do not affect protein utilization, then we can reasonably conclude that pasteurizing doses will likewise have no adverse effect. This was confirmed by additional protein-utilization studies which showed that haddock and flounder irradiated at levels up to 0.6 Mrad have the same protein

efficiency ratio as raw, nonirradiated fish.[36] It is interesting to note, however, that, although the protein quality was not adversely affected by irradiation, it was nonetheless decreased as a result of heat-treatment during cooking.

Protein-utilization studies were complemented by quantitative analyses of total and free amino acids in irradiated and nonirradiated haddock fillets and clam meats.[39,40] It was found that the amino acid content of samples irradiated at doses exceeding 2.5 Mrads was not significantly different from that of the control samples. In cases where slight changes did occur, they were not more significant than those caused by heat processing or cooking.

Some changes in water-soluble vitamins have been noted as a result of irradiation, particularly with treatment at sterilizing levels; but these changes were about the same as those which result from cooking. Losses in thiamine in salmon, lake trout, and tuna ranged from 24% at 0.2 Mrad to 94% at 4 Mrads. Haddock fillets irradiated at 2.8 Mrads lost 70% of the thiamine, but there was no change in niacin or riboflavin.[36] In tests carried out at the BCF Technological Laboratory, it was found that, with the exception of thiamine, pasteurizing doses of 0.25 Mrad or less had no significant effect on the B vitamins in haddock fillets. However, treatment at 2.5 Mrads caused slight losses of riboflavin and B-12, in addition to loss of thiamine.

Present evidence indicates that, among the water-soluble vitamins, the most radiation sensitive are thiamine, pyridoxine, and pantothenic acid; but at pasteurization doses the changes, with the exception of thiamine, do not appear significant. Thus the response of the vitamins to ionizing radiations is similar to that observed with vitamins to thermal energy for equivalent processes.

Microbiological Quality

BASIC MICROBIOLOGICAL CHANGES

Fresh Fish. No clear-cut description can be given regarding the basic microbiological changes taking place in fresh-fish fillets as they are held at refrigerator temperatures. Most of the investigations on this subject have been carried out on cod fillets. Spoilage flora and the sequence of chemical changes with this fish might be similar to those in haddock or whiting, but might be different with some other species.

In the freshly cut and/or cut and skinned cod fillet, *Pseudomonas* species appear to predominate; fairly high relative concentrations of *Achromobacter* and coryneforms also are present.[41] During storage the pseudomonads become predominant, types such as *P. perolens* first causing fruity, garbage and potatolike odors,[42,43] after which the more proteolytic pseudomonads become predominant and cause spoilage. It is indicated that for eviscerated cod held in ice, types similar to *P. fragi* constitute the major flora at the time of spoilage.[44]

The various organoleptic stages of fillets during holding at refrigerator temperatures above freezing have been characterized[45] as follows: first stage, fish characteristic odor; second stage, loss of typical odor and flavor; third stage, the development of various odors described as fruity, potatolike, etc.; fourth stage, the development of rotten, putrid, or fecal odors. These authors also suggest that trimethylamine, ammonia, hydrogen sulfide, and indole, in various concentrations, may be the cause of the fourth stage of spoilage. However, since *P. fragi* produces neither hydrogen sulfide nor indole, it would seem that other compounds are involved. It is known, of course, that various short chain fatty acids, including butyric and isovaleric, are produced during the spoilage of cod,[44] and these compounds could be partially responsible for off-odors in fillets at the time of spoilage.

Irradiated Fish. Even less is known about the spoilage of irradiated fish fillets than of untreated fish. In an investigation with cod,[46] it was found that, at the time of spoilage at 32°F, product irradiated at 0.25 or 0.5 Mrad contained a flora consisting almost entirely of pseudomonads. Other investigators[47] working with dover sole have found that in product irradiated at 0.1 to 0.3 Mrad and held at 43°F the spoilage flora consisted mainly of *Achromobacter* species. In unpublished investigations[47,48] it has been found that, in air-packed haddock irradiated at 0.15 Mrad and held at 33°F, *Achromobacter* species were predominant at spoilage.

With respect to organoleptic changes in irradiated fish fillets held at refrigerator temperatures above freezing, it has been found that, with petrale sole fillets vacuum packaged in cans, total plate counts may reach approximately 1×10^8 per gram before the radiation-

pasteurized product is judged to be unacceptable.[49] With fresh unirradiated fish of this kind, the total plate count was much lower than this at the time of spoilage.

Investigations with radiation-pasteurized haddock fillets (0.15 to 0.35 Mrad) held at 33, 35, 40, or 45°F have indicated that when the total plate count reaches the approximate level of 1×10^8 per gram, the product is already spoiled and has a putrid, ammoniacal odor.[50] Also at this time the *p*H of this product has increased from about 6.7 to about 7.5.

It would appear that, at least with some species, the spoilage flora of radiation-pasteurized refrigerated fish is different from that of the untreated product. This may partially explain why the irradiated product may be held for longer periods of time under refrigeration than can the untreated product before spoilage occurs.

ROLE OF PATHOGENS

Fresh Fish. It is known that *C. botulinum* type E may be present in fresh fish, both in saltwater and freshwater species. In waters neighboring Sweden, Johannsen[51] found that 22% of herring taken from these waters contained type E *C. botulinum* in the intestines. On the west coast of the United States, Eklund and Poysky found that in about 87% of the sole taken from the mouth of the Columbia River and up to 26% of fish taken from areas off Washington and Oregon the intestines contained type E *C. botulinum*.[52,53] The intestines of whitefish chubs taken from some areas of Lake Michigan have been found to contain type E *C. botulinum*.[54]

Very little has been done to determine the frequency of contamination of fish fillets with *C. botulinum* type E. In commercial haddock fillets it has been found that type E *C. botulinum* is present in approximately 20% of samples.[50] It is important to note, however, that this organism was usually not found in 50-g portions, and the actual concentration was estimated to be only 0.17 cell per gram at the highest level encountered. It was necessary to test 555-g samples in order to pick up demonstrable quantities of organisms.

Pathogens other than *C. botulinum* present in fish would seem to have little significance for products consumed in the United States, since fish is not eaten uncooked. Freshwater fish in certain countries may contain *Salmonella* and *Shigella*, and there have been outbreaks of staphylococcal or *C. perfringens* poisoning and salmonellosis caused by the consumption of fish.[55] However, outbreaks of this kind may have been caused by recontamination of the cooked product combined with holding at temperatures above 38°F after the fish had been cooked.

Irradiated Fish. The susceptibility of irradiated marine products to growth of *C. botulinum* type E has been important from the standpoint of public health. Since type E *C. botulinum* can grow at temperatures[56] as low as 38°F, we felt it was necessary to determine whether or not this organism is a wholesomeness problem in irradiated and nonirradiated fish.

We worked with 10-lb quantities of haddock fillets, inoculated with 1×10^2, 1×10^4, or 1×10^6 spores per gram of type E *C. botulinum*, irradiated at 150, 250, or 350 krads and held at 33, 35, 40, or 45°F.

In holding these fillets for up to 120 days at 33 and 35°F and for one, two, or three times the irradiated storage life at 40 and 45°F, toxin was found only at 45°F. In products held at 45°F, no toxin was produced in haddock inoculated with 1×10^2 spores per gram during holding for three times the irradiated storage life. Product inoculated with 1×10^4 spores per gram was toxic in three times but not in twice the irradiated storage life, and product inoculated with 1×10^6 spores per gram was toxic in one irradiated storage life.

It has also been found[50] that in radiation-sterilized haddock tissues inoculated with type E *C. botulinum* and with *P. fragi* (the latter organism contributing to the spoilage of fresh haddock) or with type E *C. botulinum* and *Achromobacter* species (the latter organism contributing to the predominant spoilage of irradiated haddock), the cultures containing *P. fragi* and *C. botulinum* type E became toxic in 10 days at 45°F, whereas the cultures containing *Achromobacter* and *Clostridium* did not become toxic until 20 to 30 days at 45°F. In either case the aerobic count at the time that toxin was detected would render the product badly spoiled organoleptically.

Considering the infrequent contamination of haddock with type E *C. botulinum* and the low level of contamination of haddock fillets with this organism, there would appear to be little problem with type E botulism. Furthermore,

uncooked haddock has been found to be a relatively poor substrate for the growth of *C. botulinum* type E, and toxin production seems to be facilitated more by the growth of bacteria-causing spoilage of fresh nonirradiated fish than by the growth of the bacteria-causing spoilage of irradiated haddock.

RADIATION-RESISTANT BACTERIA

It is interesting to compare the relative radiation resistance of the various types of organisms found in marine products as well as to compare one of the most radiation-resistant of these, *C. botulinum* type E, with other resistant species not normally associated with irradiated foodstuffs.

The bulk of the organisms present in marine products is relatively radiation sensitive; they consist mainly of pseudomonads, *Achromobacter* and other gram-negative organisms. Thornley,[57] in reviewing the data on resistance among bacteria, has pointed out that these organisms are approximately one-fiftieth as resistant as the spores of *C. botulinum*, which in the case of type A have a D_{10} value of 0.286 to 0.311 Mrad.[58,59] A similar approximate range of D_{10} value was found for type A spores by Anellis, Grecz, and Berkowitz,[60] namely, 0.295 and 0.396 Mrad, respectively, in buffer and in pork–pea broth. *Clostridium perfringens* type A spores are somewhat less resistant (0.23 to 0.30 Mrad) in phosphate buffer and in cooked meat broth.[61]

Clostridium botulinum type E spores are somewhat less resistant than the other types of *C. botulinum*, the D_{10} value being about 0.14 Mrad.[62]

Thus it would seem that one of the most resistant organisms which may be present in marine products is *C. botulinum* type E, the spores of which are much more radiation resistant than are the bulk of the organisms normally present in marine products. On the other hand, *C. botulinum* type E is much less resistant than type A spores of *Micrococcus radiodurans*, a highly radioresistant vegetative bacterium isolated in a meat-packing plant in Oregon.[63]

Fortunately surveys both of commercial fishing grounds and of the marketplace have shown type E spores to be present in low concentrations and then only in a small percentage of the samples examined.[64a]

Functional Similarity of Food-Pasteurization Processes

Radiopasteurization is similar to other pasteurization processes. Treatments with heat, chemical additives, controlled atmospheres, or combinations of these techniques, while having varying effectiveness, nonetheless generally change the microbial flora of the product in a manner so as to substantially decrease the organisms principally causing spoilage.

All pasteurization processes, including irradiation, have certain built-in safety factors that protect the public health. These include rapid spoilage at elevated temperatures and recognizable organoleptic changes that precede growth of pathogens. In addition, handling, process control, and cooking also can be considered safety features.

The concept of pasteurization is not new to the seafood industry, which has for a long period of time used heat to effect an extension of product shelf life. For example, crab meat has been handled as a fresh, cooked, refrigerated product for many years. The processes include cooking the live crabs and marketing the refrigerated cooked meat or heating the picked–cooked crab meat to an internal temperature of 170°F and marketing it under refrigeration. Since the crab meat is picked by hand, extra care must be taken to prevent recontamination.[64b]

Radiation pasteurization might be an improvement over existing crab cooking and pasteurization processes. The picked–cooked meat of Dungeness crab meat could be irradiated to eliminate or decrease any potentially harmful organisms that might be transmitted to the product during picking.

In addition to eliminating subtle quality changes resulting from exposure to high temperature, irradiation would permit more accurate control of refrigerated shelf life to ensure that organisms are present which eventually cause typical organoleptic changes.

Pasteurized hams and sausages are other examples of products that have a long-established good public-health record. Although contamination and proliferation of pathogens is always a potential likelihood, hams and sausages are processed usually containing inhibitory salts and handled in a manner so as to make any danger remote. It is interesting, however, to observe that in all cases the shelf life of

radiation-pasteurized fish fillets is much less than that for these products.

Smoking and curing must also be appraised in relation to radiation, particularly since smoked fish has been implicated in outbreaks of botulism.[55] The 1963 incidences resulted from eating smoked whitefish that are believed to have been held at ambient temperatures in the summer for as long as 5 days. Radiation pasteurization, in contrast with smoking, does not mask off-odors and flavors associated with spoilage.

DISCERNIBLE SPOILAGE

As opposed to sterilized and frozen foods, fresh produce and meat products undergo quality deterioration at a relatively fast rate.

Care is usually taken to see that the perishable products are of good quality when delivered to the consumer, but the danger of spoilage is always possible. In those rare cases where spoilage results, the consumer is warned by his ability to recognize quality deterioration by odor, appearance, or other built-in characteristics of the foods. Since spoilage of non-irradiated fish is readily apparent by smell and appearance, there is little likelihood of anyone eating products that may pose a public-health hazard. It is necessary that radiopasteurized fish and shellfish offer this same factor of safety so that spoilage, when it does occur, will be easily detected by the consumer. In this section we will attempt to show that fishery products pasteurized at low irradiation levels spoil in as recognizable a manner as fresh fish and that they possess the same degree of safety as their untreated counterpart. We will also discuss the causes of bacterial spoilage and how it manifests itself in fishery products.

Spoilage in Fresh Fish. As we discussed in an earlier section, spoilage in fish, as in other foods, is a process of quality deterioration that finally results in inedibility. The muscle of the fish is essentially sterile when alive, but soon after death the protective mechanism is believed to break down and bacteria invade the tissues and proliferate.

Since bacteria are mainly responsible for fish spoilage, the bacterial contamination of commercial fish and equipment has been given considerable attention[65,66] and the composition of fish microflora has been rather extensively studied. These studies are described by Shewan[67] who has shown that the types and numbers of bacteria that inhabit fish are affected by species, season, environment, treatment, handling, etc.

Adams, Farber, and Lerke[68,69] have presented significant data implicating *Pseudomonas* and *Achromobacter* species as the main fish-spoilage organisms.

The number of total bacteria varies mainly as a function of the holding temperature and handling practices. In commercial products the total bacterial load for fish might vary from 1×10^3 to 1×10^6 organisms per gram of flesh.

Many investigators use the total plate count (aerobic) as an index of fish quality or in conjunction with other quality indices, even though quality deterioration is a more direct function of the metabolic activity of the spoilage organisms than it is of the total bacterial numbers. This is illustrated in a paper by Hansen.[70] In fish spoilage we are, in fact, concerned with specific spoilage organisms — for example, some of the pseudomonads that produce hydrogen sulfide, an obnoxious end product of proteolysis. On the other hand, *Flavobacterium* species are not as active and produce much less offensive products than do species of *Proteus*, *Pseudomonas*, or *Achromobacter*.[71] It is clear then that, for any unit of bacterial activity, some *Pseudomonas* species can be expected to create a greater degradation of product quality than might *Flavobacterium* species. These facts need to be considered in terms of composition of the bacterial flora and any composition changes that may occur.

In general, the shelf life of very fresh fish, when they are properly iced at 33°F, is about 2 weeks. By the end of this time, the accumulation of obnoxious products resulting from bacterial activity is sufficient to make the fish definitely unacceptable though it is not necessarily harmful.

Of all tools used to evaluate fish quality, the most widely used and accepted one is the trained taste panel. While it is true that a taste panel has shortcomings, it is at least operating with the same sensory devices and is detecting by the same organoleptic criteria as do ordinary consumers. The more reliable and trained a panel is, the more confidence one can place in it. On the other hand, any of the multitude of objective tests devised and tried to date (a recent survey of these was made by Gould[72a]) lack universality, are dependent on numerous chance variables, and, at best, are only as reliable as a trained

taste panel indicates. To our knowledge there is no accepted objective test to measure the quality of irradiated fish, although recently published data[72b] indicate that the development of trimethylamine, total volatile bases, and total volatile acids is approximately the same in fish irradiated at less than 0.15 Mrad as it is in nonirradiated fish.

Spoilage in Radiopasteurized Fish. In recent years low doses of ionizing radiation (<0.5 Mrad) have been successfully used experimentally to double or triple the shelf life of many seafoods. The shelf-life extension is made possible by the destructive effect of radiation on bacteria. For example, it has been found that 0.25 Mrad reduced the total plate counts in haddock fillets by 2 to 3 log cycles.[73,74] Others have reported similar data.

Taste-panel evidence obtained at several laboratories, including the BCF Gloucester Technological Laboratory and the Nutrition and Food Science Laboratory at Massachusetts Institute of Technology, indicates that radiopasteurized fish spoil as a result of bacterial activity as do fresh fish.

In recent years, a number of studies have been carried out to determine the effect of radiation on the composition of the microflora in seafoods. Masurovsky, Voss, and Goldblith[75] found that while *Pseudomonas* was highly susceptible to radiation and was greatly reduced in numbers when irradiated at 0.05 to 0.35 Mrad, the few survivors remaining after irradiation nevertheless multiplied to about their original fraction of total bacteria during storage. *Bacillus*, *Micrococcus*, and yeasts, however, showed significant increases and became predominant. Corlett, Lee, and Sinnhuber[76] found a predominance of *Achromobacter*. *Micrococcus*, and yeasts in the survivors in sole at dose levels up to 0.30 Mrad. Sinskey et al.[77] found a drastic reduction in *Pseudomonas* species in haddock fillets irradiated at 0.15 Mrad, while *Achromobacter* increased in numbers to predominating levels. They further showed that the use of hermetically sealed packages impeded the growth of surviving aerobes, especially the few remaining pseudomonads, in a manner similar to irradiation.

Although radiopasteurization reduces the total plate counts by as much as 99.0 to 99.9%, it reduces the more biochemically active pseudomonads by even greater proportions. However, *Achromobacter* and other genera that are relatively radioresistant and any few remaining pseudomonads together are capable of spoiling irradiated fish during extended storage.

Thus accumulation of offensive flavors and odors in fresh fish occurs much more rapidly than in irradiated fish, but, although the spoilage in radiopasteurized fish is retarded, it still occurs.[78] On the basis that a change in the microbial flora is bound to create a change in the products of bacterial activity, one would assume that the spoilage patterns in irradiated fish would not only be slower but, in addition, that they would be different from those in fresh fish. However, trained panels at BCF laboratories in Gloucester, Mass., Seattle, Wash., and Ann Arbor, Mich., and other laboratories have not reported any major organoleptic differences between spoiled irradiated and spoiled nonirradiated fish at doses less than 0.15 Mrad. Therefore, whatever the differences between them, the spoilage patterns in radiopasteurized and nonirradiated fish have essentially the same effect on the organoleptic quality of the products. This is shown in Table III-6, which lists the shelf life and spoilage characteristics of irradiated and nonirradiated cod fillets stored at four different temperatures. It can be seen that the comments used by the panelists to describe the irradiated fish when it became unacceptable were much the same as those used to describe the nonirradiated fish when it became unacceptable. The major difference was that the irradiated fish required more time to reach unacceptability.

ESTABLISHED HANDLING PRACTICES AND RADIOPASTEURIZED FISH

Fresh-fish fillets have had a history of safety and wholesomeness since their introduction early in the 20th century. The industry, which is largely centered in Massachusetts, has long recognized the value of good icing, processing, and marketing practices in minimizing spoilage.

In this section we present background information on the handling and marketing of fresh-fish fillets, describe a recent BCF survey on the temperatures and conditions experienced in the marketing of fresh fish, and discuss the future distribution of radiopasteurized fish on a commercial basis. The present system of handling fresh fish is entirely adequate for radiopasteurized fish.

Handling of Fresh-Fish Fillets. In 1906, introduction of powered beam trawlers patterned

Table III-6 SPOILAGE CHARACTERISTICS AND SHELF LIFE OF IRRADIATED (0.15 Mrad) AND NONIRRADIATED COD FILLETS STORED AT DIFFERENT TEMPERATURES

Sample	Storage temp., °F	Shelf life of product served steamed,* days	Panelists' comments	
			Steamed product evaluation at end of shelf life	Raw product evaluation at end of shelf life
Irradiated in air-packed cans	33	36	Off odor, musty, old	Persistent fishy odor, slight discoloration
	37	15	Ammonia, bitterness	Opaque, defects, discolored, fishy odor
	42	12	Irradiation taste, bitter, burnt taste	Slight irradiation odor, slight fishy odor
	47	12	Ammonia, putrid, inedible	Defects and discoloration, slight irradiation odor, fishy odor
Irradiated in bulk-packed 30-lb tins	33	36	Spoiled, irradiation odor, irradiation flavor, ammonia	Slight discoloration, slight fishy odor, slight irradiation odor
	37	15	Putrid. ammonia, irradiation odor	Defects and discolored, fishy odor
	42	12	Musty, rancid	Slight fishy odor
	47	12	Ammonia, stale odor	Defects, discoloration, irradiation odor, decomposition odors
Nonirradiated, bulk-packed in 30-lb tins	33	15	Ammonia, putrid, sour, inedible	Defects and discolored, persistent fishy odor
	37	12	Ammonia, inedible, putrid	Defects and discolored
	42	12	Ammonia, putrid, rancid, sour, bad	Defects, discolored, decomposition odors
	47	5	Ammonia	

*Shelf life of deep-fat fried product is considerably longer.

after English design permitted landings of fresh iced fish. The beginning of the fresh-fish-fillet industry can be traced back to 1921, when Dana Ward of Boston first offered parchment-wrapped boneless haddock fillets for sale. Since that time the fresh-fish-fillet industry has grown considerably; and today over 80 million lb of this product are produced in the United States. In processing, fillets are cut from whole or eviscerated fish by machine or hand and then usually packed in 10-, 20-, or 30-lb tins for shipment to wholesale or retail markets. These rectangular tins, which have been used preeminently in the shipping of fillets since 1925, are quite satisfactory. The fillets are wrapped in plastic in small lots of 1 to 2 lb or are layered in bulk in the tins.

Present shipping practices for fresh fish have evolved through trial and error to the point where virtually few shipments require more than 2½ days of shipping time from point of origin to destination. This affords enough time for the fish to be sold and consumed in a reasonably good fresh state. A BCF survey of distribution practices of the fresh-fish industry showed that commercial conditions of transportation are not detrimental to the safe distribution of fresh fishery products.

The distribution of fresh fillets from producer to ultimate consumer has varied insignificantly ever since the inception of commercial filleting in 1922. The fillet shipping containers, whether rectangular metal tins or heavily wax-coated fiberboard, are embedded in ice in wooden barrels or boxes and shipped via truck or rail. Depending on the distance the fillets are to be shipped, the trucks may be equipped with mechanical refrigeration; the longer the trip, the greater the probability that the truck will be insulated and will use mechanical refrigeration. Barrel shipments made by railroad to distant points are reiced once in transit or as needed. Railway cars may or may not be insulated and refrigerated and may also carry other types of nonperishable merchandise with seafoods. The weight of ice per barrel usually equals or even exceeds the weight of the fillets. While simple and almost primitive in operation, it is very effective in maintaining fish-flesh temperatures close to that of melting ice.

The largest amount of fresh fish sold at retail is through chain stores. None of the chain stores, however, attempted to sell fresh fillets until 1925—3 years after small independent stores had recognized the value of fish in fillet form. From the onset, retailers sold

unbranded bulk layer-packed fillets. Branded fresh fillets that consisted of fillets weighing about three-quarters to approximately a pound first made their debut in 1922.

The retail method of holding fillets has generally been to place them on or in ice. Some of the more modern retail display cabinets use a combination of ice and mechanical refrigeration, and others depend almost exclusively on mechanical refrigeration. Internal flesh temperatures of fresh fishery products held in retail dispensing cabinets run from 34 to 46 °F.

With the current emphasis upon packaging foods, many retail stores, particularly the chain stores, sell most of the seafood items in individual consumer-sized units of varying weights. The most common practice is to place a certain amount of fish or fillets in cardboard trays and overwrap with a clear plastic film and a sticker bearing species identification, weight, and price. The common cardboard trays in a very few areas are now being replaced by lightweight plastic trays which present a neater appearance when overwrapped with a plastic film which is tightly sealed to prevent loss of free liquid. A widespread practice in many of the supermarket stores after closing Friday night is to half the original retail price of displayed packaged fish for quick sale on Saturday in order to reduce waste.

Survey of Fresh-Fish Distribution Practices. With the advent of commercial irradiation of fishery products near, we considered it necessary to learn as much as possible about the most recent commercial practices of fresh-fish distribution. Information of this nature is essential to determine just how suitable present practices are for the safe distribution of irradiated fishery products. To obtain details on shipping and marketing fresh fish, we conducted a survey of the New England industry.

The purpose of the survey was to obtain and record temperature of fresh fishery products in the entire chain of distribution from fish pier to inland consignee and ultimately to the retail display case. This was done by sending an observer with the interstate truck or railway car to record temperatures and observe conditions of handling, delays, and possible abuses at all stages of the trip. Samples of nonirradiated and irradiated fillets of identical history were compared to determine differences and their acceptability by a trained taste panel at the BCF Technological Laboratory.

The survey lasted for a year so as to include shipments (usually two) in each of the four seasons of the year. Shipments left the Boston or Gloucester areas via licensed interstate, insulated, refrigerated trucks or regular railway freight cars to inland destinations representing the usual extreme distances to which fresh fish are normally distributed. Areas chosen were Cleveland, Ohio, with a run of 66 hr and 660 miles; Miami, Fla., with a run of 5 days and a distance of 1550 miles; Texarkana, Tex., via rail with 2 days' travel time and 1715 miles; and Seattle, Wash. (one railway trip only) with 5 days' travel time and 3159 miles. In addition to these, several trips were made in privately owned trucks by a processor who did his own distribution in states contiguous to Massachusetts.

The processor–distributor trips were undertaken to study short hauls that included many stops for drop shipments. Truck doors were opened for as many as 28 deliveries. There was little or no change in product temperature from point of origin to destination.

The temperatures of all shipments made by common carriers were always effectively lowered several degrees by the time of arrival at destination. The products on the Florida run in some instances were actually partially frozen because of long running periods of the refrigeration units. Temperatures at destination were effectively lowered to or near the freezing point.

This survey, which is still in progress, indicates that present distribution practices in the fresh-fish industry result in the safe distribution of fresh fishery products. While abuses have been observed, they are common to the distribution of all types of perishable foods. There were no particular hazards to fresh fishery products whether irradiated or not.

Handling Radiopasteurized Fish Fillets. Radiopasteurized fish fillets probably will be packaged and distributed using the same well-established practices that have been in use for fresh fish. The fish will be irradiated at dose levels seldom exceeding 0.2 Mrad. Cellophane, polyethylene, and wax-coated paperboard account for most of the packaging materials in direct contact with fish. Fillets to be radiopasteurized will be packaged in one of three

ways. Twenty to thirty pounds of bulk unbranded fillets will be placed in a polyethylene bag inside a metal fillet tin or wax-coated paperboard box and then irradiated. Branded fillets will be treated as conventionally loosely hand-wrapped fillets in approved film or in a tray overwrapped tightly by a heat-sealable transparent film. Some types of fillets or fish steaks may be wrapped in a heat-sealable bag of approved film. Irradiation will follow packaging regardless of the style of finished package.

Shipping practices for radiopasteurized fish will follow those currently in use for nonpasteurized fresh fish. The present practices are so geared as to permit a limited shelf life of seafoods at refrigerated temperatures. In general, the shelf life of fresh nonpasteurized fish is about 1 week at the time it leaves the dealer's premises. At the retail level, there are usually about 3 days of good shelf life left; but after this period, the quality decreases rapidly.

Radiopasteurized fish will be sold by retailers in essentially the same fashion as fresh nonirradiated fish. Bulk fillets enclosed in a polyethylene or other approved film bag and irradiated in a metal or waxed fiberboard container will lose their integrity once they are opened for sale at retail. They will, however, have a longer shelf life than nonirradiated fillets because of the protection afforded by radiation and packaging against recontamination up to the point at retail where the protection is lost.

Bulk irradiated fillets once opened by the retailer will have a shelf life of about a full week as compared to only 3 days for nonirradiated fillets. Individually packaged fillets will have a shelf life of 10 or more days provided package integrity has been maintained.

Normal decomposition of both bulk and packaged irradiated fillets will occur when limits of shelf life have been exceeded.

COOKING

In the United States, in contrast to some other countries such as Japan, fish fillets are consumed in the cooked state. This custom can be regarded as a safety factor against botulism whenever the potential for this type of food intoxication is present, because of the heat-labile nature of the toxin.

Studies have been conducted to determine the adequacy of the cooking process on the inactivation of type E botulinum toxin in haddock fillets.[50] A thermal destruction time curve was determined, first of all, for the toxin at a concentration of 10,000 LD_{50} mouse units per milliliter in a raw-haddock substrate. It was found, for instance, that between 50 to 60 min at 140°F or 2 to 2.5 min at 160°F were required to inactivate the toxin. Next, the rate of heat penetration into a $\frac{5}{8}$-in.-thick haddock fillet was determined during deep-fat frying at 375°F by means of a thermocouple located at the center of the fillet. The fillets were cooked for 4 min and then allowed to air-cool, in accordance with commercial practices. A temperature of about 150°F was attained at the center of the fillet at the end of the 4-min cooking; however, the temperature continued to rise when the fillet was removed from the hot oil, reached a maximum of 172°F by 7 min, remained at this temperature until about the tenth minute, and then very slowly diminished. The lethality curve for the cooking process was constructed from the heat-penetration data and thermal destruction time curve. By graphical integration of the area under the curve, it was calculated that the toxin, at a concentration of 10,000 LD_{50} mouse units per gram of haddock, would be inactivated in 5 min, that is, 4 min of cooking plus 1 min of air cooling. Since it is very unlikely that the fish would be consumed immediately after removal from the hot oil, but rather after a 5- to 10-min delay, due to a variety of reasons, then the time lag from cooking to eating represents a safety factor for toxin destruction.

In this study the thermal resistance of the toxin was determined at a *p*H of 6.7, which was the *p*H of the fresh haddock. Ohye and Scott[79] observed that the heat resistance of type E toxin is strongly *p*H dependent, being most resistant at about *p*H 5. It has been shown that when radiopasteurized haddock fillets, which were inoculated with type E botulinum spores, spoil at refrigerator storage temperature, the *p*H of the flesh is alkaline. Thus, in reality, type E botulinum toxin would probably be inactivated faster during the cooking process than at the experimentally calculated rate, since the latter was determined under less adverse *p*H conditions for toxin stability than would probably exist if and when type E toxin were produced in a haddock fillet.

Further studies with inoculated packs of radiopasteurized haddock fillets (inoculated with 1000 spores/g) showed that the maximum toxin titer formed at a storage temperature of 45°F was 100 LD_{50} mouse units per gram, and this occurred after the sixth week. However, the fillets were regarded as decomposed after 4 weeks, at which time the total plate count was about 800×10^6 spores per gram. At a storage temperature of 75°F, the maximum toxin concentration of 10,000 LD_{50} mouse units per gram was reached at the end of 7 days. However, the fillets were regarded as decomposed after 3 days, at which time the total plate count was approximately 300×10^6 per gram. The toxin titer at this time was 100 LD_{50} mouse units per gram.

A relation between thermal inactivation time and toxin concentration was found to exist, such that for a 10-fold decrease in toxin concentration the thermal inactivation time decreased by a factor of 5.5. On the assumption that a spoiled haddock fillet would only contain a toxin titer of 100 LD_{50} mouse units per gram, recalculation of the original data indicates that this level of toxin in a $\frac{5}{8}$-in.-thick fillet would be inactivated after 3.5 min of deep-fat frying.

Abuse of the product by holding at above refrigerator temperatures would not be expected to present any additional hazard because under these conditions, growth of the surviving microflora would also be favored, and this would lead to the production of fruity, fishy, and ammoniacal odors signaling spoilage. As was indicated previously, when radiopasteurized inoculated haddock fillets were held at 75°F (and this can be considered an abuse), the fillets were not toxic after 2 days of storage, but after 3 days of storage the total plate count was about 300×10^6 per gram, an ammoniacal odor was evident, and type E toxin was present at a concentration of 100 LD_{50} mouse units per gram.

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Section

IV

Isotopes and Radiation Technology

Applications in Medicine

The use of radioisotopes in human medicine for both diagnosis and therapy is well known, but their use in veterinary medicine is less publicized. For both the research worker and the veterinarian, radioisotopes can be a useful tool. A few of the techniques that have been investigated at the New York State Veterinary College are described here.

Radioisotopes in Veterinary Medicine*

By F. A. Kallfelz and R. H. Wasserman†

A few of the uses of radioisotopes that have been investigated at the New York State Veterinary College are reviewed here. Diagnostic uses include ^{131}I for determining thyroid and liver function, ^{59}Fe for determining parasitic blood loss, and ^{203}Hg for locating brain tumors; therapeutic uses include ^{90}Sr for treating tumors.

The use of radioisotopes for both diagnosis and therapy of animal conditions has been investigated at the New York State Veterinary College.

The use of atomic energy in veterinary medicine still has many areas into which it can expand. Most of the work with radioisotopes is confined to veterinary colleges or large-animal hospitals where practitioners may refer cases for these specialized procedures. In the future, however, many of these techniques will be more readily available to the individual practitioner to help him better serve the needs of his clients.

*Reprinted, except for the abstract and part of the first two paragraphs, from *Farm Research*, 33(3): 6-7 (1964), by permission of the author.

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Thyroid Function Test

Radioactive ^{131}I is used in veterinary medicine for determining the functional state of the thyroid gland, which is important in the differential diagnosis of many hormonal disturbances, tumors, and mineral deficiencies. The test is easy to perform, and the animal suffers no ill effects. A small dose of radioactive iodine (an element preferentially accumulated by the thyroid gland) is administered intravenously. After 72 hr a radiation detector placed at a specified distance from the animal's neck measures the amount of iodine absorbed by the gland. Thus it can be determined whether the gland is normal, overactive, or underactive.

Another indication of efficiency of thyroid function is the rate of conversion of inorganic radioiodine to the protein-bound form. For this test a sample of blood plasma is obtained 72 hr after the injection of the isotope and passed through an ion-exchange resin which removes inorganic iodine. By measuring the radioactivity of the plasma both before and after its passage through the column, the percentage of the radioiodine converted to the protein-bound form can be determined. The greater the amount of conversion, the more active is the thyroid gland.

Liver Function Test

One of the many biochemical tests devised to check the functional status of the liver for clinical signs of disease uses a drug called "Rose Bengal." The liver has the ability to remove this drug from the bloodstream, and the rate of removal is directly related to its functional status. Labeling of Rose Bengal with ^{131}I has made this test more precise in that uptake and excretion of the drug by the liver can now be detected with an externally placed

radiation detector, which obviates the tedious colorimetric analysis of plasma (Fig. IV-1). This test may well help to shed light on such diseases as ketosis in cattle and pregnancy toxemia in sheep, where liver dysfunction is assumed but not proved.

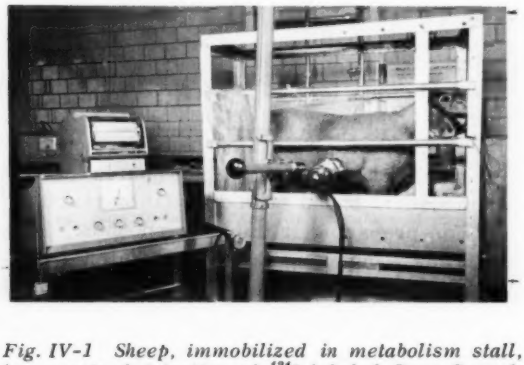

Fig. IV-1 Sheep, immobilized in metabolism stall, has received injection of ^{131}I-labeled Rose Bengal. Uptake and excretion of drug by liver cells is monitored by means of external radiation detector focused on region of the liver. This will indicate whether liver is functioning properly.

Determination of Parasitic Blood Loss

By use of radioactive iron (^{59}Fe) and a whole-body counting technique, it is now possible to estimate the amount of blood loss due to blood-sucking intestinal parasites in sheep. When radioactive iron is injected into a sheep, it becomes incorporated into the red blood cells. The radioactivity of the sheep is counted periodically in the large counter, and the rate of red cell loss is determined. The more rapid the rate of loss of red cells, the more severe the parasitic infection. Better recommendations for the control and therapy of intestinal parasitisms result from such studies.

Brain Scanning

Diagnosis of certain intracranial lesions, such as brain tumors, is facilitated with isotopic tracers. A compound that is concentrated by the tumor is labeled with a radioisotope and is administered intravenously to the anesthetized animal. After allowing sufficient time for concentration in the tumor, the animal's head is placed under an automatic scanning radiation detector, which produces a printed picture of the regions of isotopic concentration, or tumor sites (Fig. IV-2). This information can be of great value in the differential diagnosis and treatment of central-nervous-system disorders.

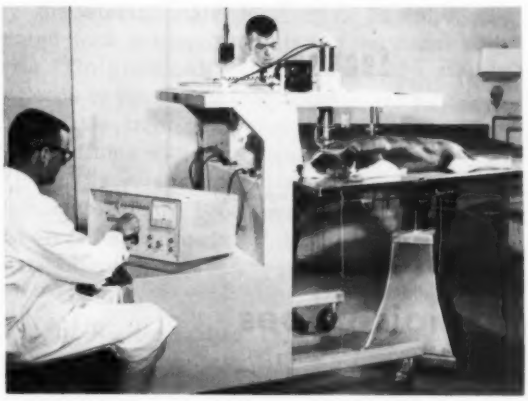

Fig. IV-2 Localization of Neohydrin labeled with ^{203}Hg, by use of scintiscanning device, in brain of anesthetized dog. This is a diagnostic test for locating brain tumors.

Beta-Ray Applicator

Unlike the above diagnostic tests, radioisotopes in high concentrations and in sealed containers have been used to deliver large doses of beta radiation to selected sites on or near the surface of the body. A ^{90}Sr probe, for example, can be used in the beneficial treatment of certain types of tumors of domestic animals (Fig. IV-3). Beta rays penetrate deep enough to destroy the tumor tissue but not so deep as to damage internal organs.

Fig. IV-3 Cow being treated with beta rays for tumor of the eye. The beta-ray source, ^{90}Sr, is concentrated in sealed container at applicator tip.

Section

V

Isotopes and Radiation Technology

Isotopes Around the World

Radioisotopes in Romania[1]

The burgeoning industry of Romania[2] has included the use of isotopes in its expansion: many isotopes and labeled compounds are produced; a large-scale source is in use for the manufacture of chemicals; small sources are used in gages; and modification of agricultural products, as well as crop protection from insects, by the use of radiation is under study. Radioisotope tracers are used in chemical analysis and research, industry, hydrology, medicine, agriculture, and forestry. Some of the more significant developments are summarized here.

Preparation of Radioisotopes and Labeled Compounds[1a]

In 1963 Romania produced 42 different radioisotopes, with a total radioactivity of 35 curies. For two of these, radioactive iron and sulfur, new production methods have been developed.

In developing the method for iron, which is based on the Szilard–Chalmers reaction, various iron complexes were irradiated in the reactor. Of these, only $K_4Fe(CN)_6$ and $K_3Fe(CN)_6$ were not decomposed, and both recovery and specific activity were highest with the ferrocyanide. The iron is recovered from the target material by precipitation with ammonium hydroxide, extraction with anhydrous ether saturated with HCl, and cation exchange.

In the new method[3a] for ^{35}S, an HCl target is irradiated and then heated to 500 to 900°C, and the product is extracted as the element by trichloroethylene. In an earlier method, ^{35}S was separated from reactor-irradiated KCl on activated alumina, eluted, and purified by cation exchange.[3b]

New methods have also been developed for ^{14}C labeling of aromatic compounds in the ring, by a method based on a diacylation reaction,[4a] and for preparing tritiated compounds.[4b,c] The tritiating agent is reactor-irradiated lithium aluminum hydride in which the ^3H formed by the $^6Li(n,\alpha)$ reaction has replaced some of the hydrogen.

Radiation Sources

CHEMICALS MANUFACTURE[1b]

A process was developed for using the gamma radiation from a 100-curie ^{60}Co source to initiate the reaction in the oxidation of technical-grade paraffin to produce fatty acids. The Bucharest chemical plant that now uses this process formerly used potassium permanganate to initiate the reaction. The economies introduced by elimination of the catalyst, together with the simplification of the technology made possible by the absence of slime and emulsions, resulted in a 20% cut in the price of fatty acids.

In this process (Fig. V-1), raw paraffin is fed to the irradiation chamber. After being irradiated, the paraffin goes to the oxidizer and is oxidized with air until a maximum peroxide value is reached. The oxidized product is converted to fatty acids, while the unsaponified product is recycled to the feed. At the start of a run, the oxidizer is loaded with a mixture of 96% technical-grade paraffin plus recycled unsaponifiables and 4% paraffin peroxides. After each run some of the product is stored in the peroxide chamber for use in the subsequent run.

MEASURING INSTRUMENTS[1b]

Radiometric equipment designed and manufactured includes gamma–gamma densitome-

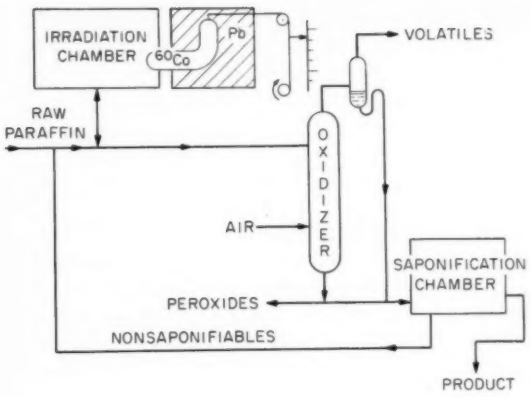

Fig. V-1 Romanian process for oxidation of paraffin with ^{60}Co used to initiate the reaction.

ters with portable counting devices. These instruments are used for measuring the density of soil on which heavy structures, such as blast furnaces, are to be built; of filler material for dikes and dams; of concrete in road building; and even of house foundations. A humidity meter based on determination of the thermalization of fast neutrons from a radium–beryllium source is used for both surface and subsurface moisture measurements. Level gages have been adopted by many industrial firms; in 1964, 50 such gages were installed by one chemical concern alone.

Gamma radiography is widely used on building sites of large industry and in construction of gas lines. This application is possible because of the mobile irradiation units made available by the Bucharest Institute of Atomic Physics.

AGRICULTURE AND FORESTRY[1e]

Seeds of wheat, barley, oats, flax, and corn, which were irradiated with X-radiation and thermal neutrons at Brookhaven National Laboratory, USA, have been field tested. Strains of wheat and barley have been found which are 6 to 24% more productive than the original. Corn with two or three fertile ears and barley which matures 5 to 10 days earlier and which is more resistant to disease than normal barley have been found. Soya seeds irradiated to 303 r with ^{60}Co gamma radiation had 5.8% more protein than normal, and those irradiated to 1925 r had 3.6% more fat.

In oak forests the moth *Lymantria dispar* L., which defoliates the trees, has been controlled by the sterile-male technique. Females that mated with males irradiated in the pupal stage with ^{60}Co produced eggs that were 85% sterile.

Seeds of the linden tree showed 31% germination in the first year when they had been irradiated either with a ^{60}Co gamma source or with ^{32}P or ^{131}I in solution. Untreated seeds did not germinate at all. Biochemical analyses showed the irradiated seeds to have a more active metabolism.

Use of Radioactive Tracers

CHEMICAL ANALYSIS[1a]

Titration. A significant Romanian contribution to the field of analytical chemistry was the development[5] of a radiometric titration method using complexing reagents that do not form precipitates, change the color of the medium, or form extractable complexes. The reaction takes place in a two-phase system: A cation in solution is titrated with a complexing anion solution under conditions so chosen that a labeled solid indicator present does not dissolve until the cation has reacted completely. For example, Ca^{2+}, Sr^{2+}, or Mg^{2+} may be titrated with ethylenediaminetetraacetic acid (EDTA), the end point being reached when solid $^{110m}AgIO_3$ dissolves:

$$^{110m}AgIO_3 + (H_2Y)^{2-} \rightleftharpoons (H^{110m}AgY)^{2-} + IO_3^- + H^+$$

Alkaloids may be determined by radiometric titration of their chlorides, sulfates, or phosphates with silver nitrate solution labeled with ^{110m}Ag. In amounts of 10^{-4} to 10^{-5} g, the error is less than 1%. Strychnine has been similarly determined by titration with Meyer reagent (K_2HgI_4) labeled with ^{203}Hg. Other compounds to which the method has been applied are cyanides, oxalates, and sulfamides.

Gravimetric Methods. The sensitivity of gravimetric methods has been greatly increased by labeling the reagent. For example, in the gravimetric determination of beryllium and bismuth, the cobalt hexammine complex is formed. If the reagent is labeled with ^{60}Co, the limit of detection becomes 10 μg of beryllium (±5% error), compared to 1.6 mg with the unlabeled reagent. Similarly, 0.08 mg of mercury may be determined as $HgI_4 \cdot Cuam_3$ (where "am" represents ethylenediamine or propylenediamine) when the iodine is labeled with ^{131}I (±5% error).

Radioactivation Analysis. Impurities in ultrapure silicon have been determined by radioactivation analysis, as have hafnium and rare earths in minerals. A radioactivation analysis method was developed for determining the amount of gold in concentrates and of losses in refining-plant tailings. Boron in minerals has been determined by the $^{10}B(n,\alpha)^{7}Li$ reaction, using a polonium–beryllium source with a flux of 2×10^5 n/sec. Gamma spectrometry has been used to determine the gold content of very-low-grade ores.

STUDY OF PHYSICAL AND CHEMICAL REACTIONS[1a]

The structure and reactions of numerous inorganic and organic compounds have been studied with radioisotopes.

Mechanisms investigated include solvent extraction, metal diffusion, and ion exchange.

CHEMICAL INDUSTRY[1b]

The use of radioactive tracers for solving chemical technology problems has resulted in process improvements with attendant economic benefits. For example, in the manufacture of superphosphate from apatite, conditions for mixing, granulating, and grain drying were determined with ^{32}P-labeled phosphoric acid. Selected as process conditions from the data obtained were: a mixing time of 1 to 6 min, depending on the raw material used; a granulation time of 6 min; a drying tray inclination of 5°; a feed rate of 11 to 12 tons/hr; and 12% moisture content. Two plants have greatly improved their processes as a result of these findings.

The source of abnormal mercury losses in a sodium chloride electrolysis plant using Krebs cells was found with ^{203}Hg-labeled mercury. The amount of this tracer in the hydrogen off-gas from a cell showed that inefficient hydrogen washing was causing the loss. Replacement of the washer with an improved type resulted in 90 to 95% mercury recovery, with a corresponding decrease in the specific consumption of mercury.

The cause of anomalies in operation of a carbide-manufacturing furnace was found with ^{32}P. The vertical cylindrical furnace has three Söderberg electrodes at the points of a triangle but drainage at only two places (Fig. V-2). It is fed with a mixture of calcium oxide and coal

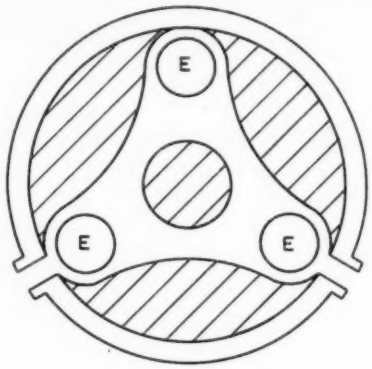

Fig. V-2 Cross section of carbide-manufacturing furnace showing electrodes (E). Hatched sections indicate solid material.

at about 2000°C. Introducing the isotope, as $Ca_3(PO_4)_2$ in the feed, at various places and counting the activity of the outlet streams indicated that a central plug of solid material was causing some of the erratic results.

PETROLEUM INDUSTRY[1b]

The development of radiotracer applications in the petroleum industry is the normal consequence of the high efficiency and applicability of radioisotopes in this field and of the importance of this industry in the Romanian economy. Technological problems can be solved with radiotracers in a tenth of the time required by standard methods, and wells are thus quickly returned to normal production.

Studies on increasing the efficiency of hydraulic fracturing have leaned heavily on data supplied by radiotracer operations. For example, labeled naphthalene pellets, rubber balls, or cardboard chips are injected, and the hydraulic fracturing operation is observed by gamma logging. The data indicate the progress of the operation.

The flooding pattern of water injection into a deposit is followed by labeling the water with isotopes; ^{65}Zn, ^{131}I, and ^{2}H are used simultaneously. Subsequent determinations of the isotopes at other locations gives data from which the movement of the water can be plotted.

MINING INDUSTRY[1b]

Ore-processing parameters were determined, for the first time in Romanian gold mining, by the use of ^{198}Au. These included ore-slurry

circulation speeds, flotation and cyaniding times, gold grain-size distribution, and losses in the barren streams and slags. The tracer was used in the form of metal alone or fastened on quartz.

PHARMACEUTICAL INDUSTRY[1b]

Codeine labeled with ^{14}C has been used to determine losses in the preparation of codeine by methylation of morphine. When the codeine separation step was shown to include a series of operations detrimental to quality and efficiency, the process stream was simplified, with an increase in efficiency from 78 to 92% and a decrease in process time and in raw-material consumption.

Similar studies with ^{14}C-glutamic acid on the preparation of glutamic acid by casein hydrolysis may result in process improvements.

HYDROLOGY[1c,6]

Radioisotopes are used for determining the flow of underground water that supplies drinking, industrial, and irrigation services and for determining drainage, infiltration, and requirements for foundations.

An example of the type of flow measurements is a test that was made in the Black Sea region, where the soil is sandy but rich in organic matter. In order to find out if a freshwater lake in the vicinity was being polluted with industrial waste from a nearby factory, the subterranean water was tested at three places, 800 m apart, on the 200-m-wide beach. At each spot, into a central well, several liters of a solution containing $^{24}NaCl$ and $^{140}La(NO_3)_3$ was introduced about 1 m below the surface of the subterranean water; the radioactivity was detected, with Geiger counters, in other wells 1 to 5 m away. The results, which showed the underground water flow to be in a north-to-south direction at a rate of 0.6 m/day, reversed previously held ideas about drainage in the region. The radioactivity in the water decreased to tolerable levels for ^{24}Na in 7 days and for ^{140}La in 30 days.

An interesting method was used to detect leaks in a water conduit under pressure. A community whose water was delivered by a concrete pipe wished to increase its rate of potable-water supply but was not sure if existing conduits could withstand the increased pressure. The system was tested by first sending a ^{24}Na solution through the pipe under high pressure (Fig. V-3) and then draining the system and drawing Geiger counters through the pipe. When the counter was drawn through, two peaks appeared on the activity record, showing two points where the radioactive solution had been forced out into the soil through weak places in the pipe.

AGRICULTURE AND FORESTRY[1e]

The availability to plants of phosphorus in the soil has been studied with ^{32}P. The form in which the phosphorus is applied, the presence of mineral fertilizers and fixation bacteria, and the nature of the soil have been shown to affect the assimilation of the phosphorus. This tracer has also been used to show the effect of the growth hormone gibberellin on the composition of bean plants.

Optimum conditions for growing fir and poplar trees and the best methods for applying insecticides have been determined with ^{32}P. The tracers ^{42}K (as chloride) and ^{86}Rb (as carbonate) have been used to study circulation of sap in oak trees.

MEDICINE[1d]

Radioactive isotopes produced at the Bucharest Institute of Atomic Physics are widely used in Romania in biology and medicine. At the first Romanian conference of specialists in this field, held in 1962, more than 100 pertinent papers were presented.[7] Papers reporting original work and isotope uses are published in both Romanian and foreign journals.

Studies in endocrinology using ^{131}I have been directed particularly toward the physiology of the thyroid hormones, diagnosis of hyperthyroidism, and clinical and experimental physiopathology of the thyroid. Investigators have determined the hormone present in the body outside the thyroid gland by scanning the thigh or forearm after oral administration of ^{131}I. Among the subjects that have been investigated are extrathyroid fixation of iodine and the effect on it of methylthiouracil, the correlation of ^{131}I values in the thigh with iodine uptake by the thyroid 2 hr after ^{131}I administration, and the uptake of radioactive thyroxin by the lung.

The isotope ^{32}P has been used to study phosphorus metabolism in animals with experimentally produced tumors. Bone metabolism, nucleic acid turnover in various organs, and

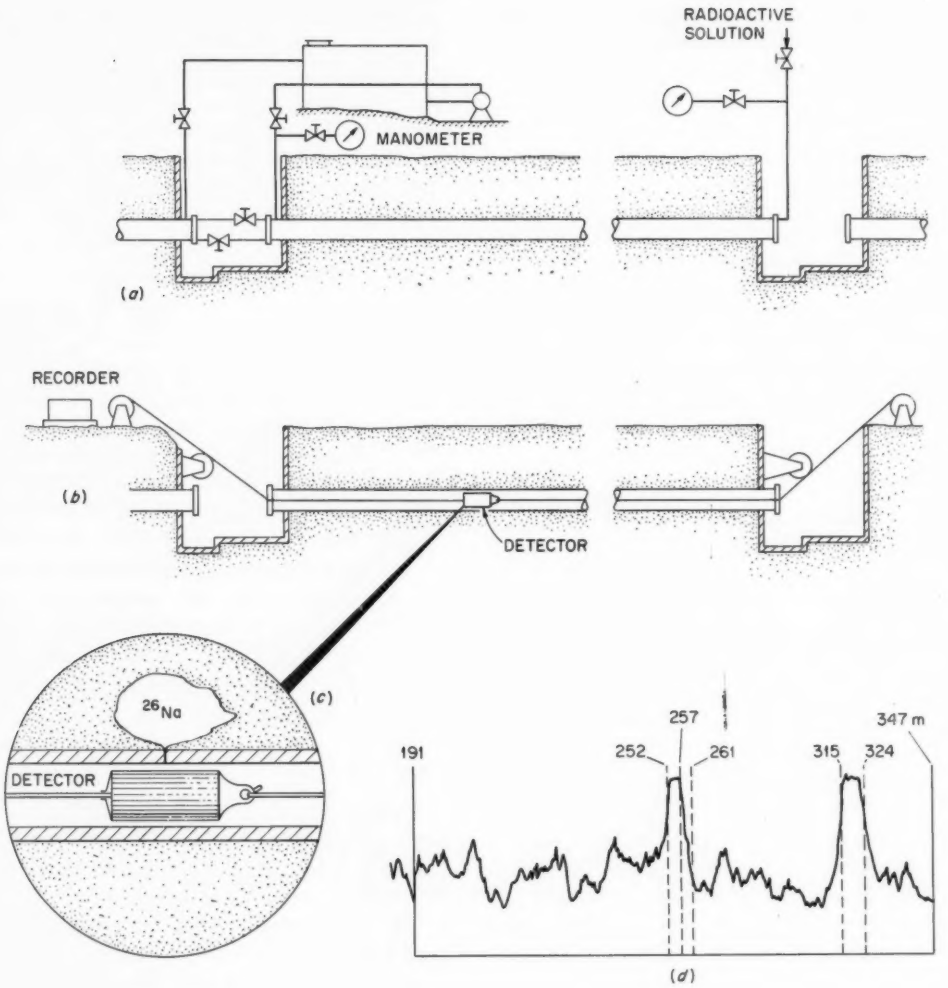

Fig. V-3 System for testing leaks in pipes with radioisotopes. (a) *Pipe through which radioactive solution is flowing.* (b) *Pipe with detectors passing through.* (c) *Enlargement at position of leak.* (d) *Record of activity, showing two leaks.*

various effects of ionizing radiation have also been studied with ^{32}P. Clinical diagnoses of malignant tumors have been histologically correlated with ^{32}P diagnoses in 82% of the cases.

Radioactive iron has been used in the study of transferrin, a beta globulin in the blood that carries iron to the tissues. Both ^{55}Fe and ^{59}Fe have been used to show that structural differences in transferrin molecules do not affect their function. Biochemical changes in erythrocytes have been shown with ^{32}P.

In studies on the gastrointestinal tract, absorption of fats has been investigated with fats labeled with ^{131}I. Both ^{35}S-labeled methionine and colloidal ^{198}Au have been used to study liver function. In colored scintigrams, different levels of radioactivity are shown by the different colors. (Martha Gerrard)

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Section VI

Isotopes and Radiation Technology

Miscellaneous Items of Interest

Conferences

Preparing, Storing Labeled Compounds Subject of Euratom Brussels Meeting

Euratom will sponsor the second international conference on methods of preparing and storing labeled compounds to be held on Nov. 28–Dec. 3, 1966, at Brussels. This conference had been scheduled for June 6–11, 1966.

A preliminary program of the conference is given below:

Lectures

H. Lettre	Recent Results in Cancer Research Using Labeled Compounds
D. Rittenberg	Historical Development of the Field of the Synthesis and Analysis of Stable Isotopes

Reviews on Preparation Methods and Tracer Applications

J. L. Garnett	Recent Developments in Heterogeneous Catalytic Labeling with ^{2}H and ^{3}H
A. P. Wolf	Radiochemical Methods of Labeling
R. Wolfgang	Mechanism and Intrinsic Limitation of ^{3}H Labeling
R. Paoletti	The Use of Drugs in Biosynthetic Preparation of Labeled Sterols and Sterol Precursors
H. K. Mangold	Synthesis and Biosynthesis of Labeled Lipids
R. Cardinaud	Nucleic Acids — Nucleotides and Nucleosides Biosynthetically Labeled
L. Pichat	Some Recent Chemical Synthesis of Labeled Compounds
A. Balaban	Intramolecular Isotopic Rearrangements of Carbon Atoms
J. G. Burr	Tracer Applications in Organic Radiation Chemistry

CHEMICAL SYNTHESIS

Methods improved: Reduction of carboxyacids by $LiAlH_4$; new homogeneous catalytic techniques; tritioboration labeling; exchange reaction of nucleosides with ^{32}P; methylene insertion reactions; use of vinyl magnesium bromide; transcarboxylation in presence of $^{14}CO_2$; Wittig reaction; Amadori rearrangement; catalytic tritiation ($AlCl_3$).

Compounds prepared: ^{14}C — Adrenaline (ring); azacholesterol and cholestane derivatives; L-asparagine; L-glutamine; glutathione; DL-hydroxyproline and DL-allohydroxyproline; cyclic ketones; DL-aspartic acid; DL-diaminobutyric acids; DL-homoserine; monovinylacetylene and divinylacetylene; α,β-unsaturated and hydroxypalmitic acids; etc.

High-specific-activity ^{14}C compounds — Bipyridilium salts; Ph-pyrimidine; Me-Ph-S-triazine; α,β-deoxyadenosine; cyclopentadiene; benzene; butanol; isohexanol; aminopropanol; etc.

^{3}H — Noradrenaline; phenoxybenzamide; cyclohexene; metanephrine; morphine derivatives; α-methyl DOPA; α-methyltyrosine and analogs; etc.

$^{125 \text{ or } 131}I$ — ^{131}I-*p*-Iodohippuran; $^{125 \text{ or } 131}I$-iodometanephrine; ^{131}I-labeled oils; $^{125 \text{ or } 131}I$-triolein; $^{125 \text{ or } 131}I$-oleic acid; etc.

^{32}P — Parathione and other insecticides; nucleotides and oligonucleotides; etc.

^{13}C — Propene; 1, 2, ^{13}C-tetradeuteriobenzene; etc.

^{2}H — α-carboxylic acids; linear fatty acids; trimethylamine alkynes; diphenylacetylene; methylaromatic acids; heterocyclic compounds; formaldehyde; etc.

RADIOCHEMICAL SYNTHESIS

Labeling by $^{197,203}Hg$ using ion exchange; labeling by exchange with carrier-free ^{32}P; radiation-induced exchange with halide isotopes; radiation-induced tritium exchange; tritium-exchange labeling of steroids and bile

pigments; polystyrene exchange tritiation; exchange tritiation of protein; recoil labeling of mercury and bromine compounds; tritiation by the use of $LiAlH_4$.

BIOCHEMICAL SYNTHESIS

^{59}Fe-ferritin from grown HeLa cells; photosynthesis of high-specific-activity ^{3}H carbohydrates; high-specific-activity nucleotides; amino acids and sugars from grown Chlorella; ^{14}C humic acid formation in soil; ^{14}C, ^{3}H, ^{35}S and ^{15}N labeling of S-adenosylmethionine from yeast culture; ^{32}P labeling of plasma phospholipoprotein.

ASSAYING

Tritium scanning; application of solubility phase equilibrium to purification of high-specific-activity complex compounds; routine purity of compounds labeled with short-lived isotopes; electron microscopic investigation of autoradiographs; counting of pure $^{14}CO_2$ in the proportional region; measurements of ^{3}H after conversion to ammonia; degradation by pyrolitic radiogas chromatography; etc.

STABILITY AND STORAGE

^{3}H thymidine; ^{3}H hexoestrol; $^{125,131}I$-thyroxine; high-specific-activity phenoxybenzamine; etc.

Large-Scale Production, Applications of Radioisotopes Discussed in Georgia

An American Nuclear Society topical meeting on large-scale production and applications of radioisotopes was held on Mar. 20–23, 1966, at Augusta, Ga.

Proceedings of this meeting, which was summarized recently by *Nuclear News*, 9(5): 17–18 (May 1966), are in press. The document, DP-1066, will be available from Clearinghouse for Federal Scientific and Technical Information, Springfield, Va. 22151, for $7.00.

AEC Activities

Nuclear Battery for Lighthouse Moved from Chesapeake Bay to Gulf of Mexico

A 60-watt nuclear generator that has been powering an unmanned beacon in the U. S. Coast Guard's Baltimore Lighthouse in Chesapeake Bay since May 20, 1964, was removed Apr. 21, 1966, for transfer to an offshore oil and gas platform in the Gulf of Mexico.

The generator will replace an identical one that was installed in the Gulf on June 21, 1965, but which, for undetermined reasons, experienced a sharp decrease in power. [See *Isotopes and Radiation Technology*, 3(1): 78 (Fall 1965).]

DID Technical Director Appointed

E. E. Fowler, director of AEC's Division of Isotopes Development (DID), recently named W. E. Mott as assistant director for technical programs.

Mott formerly was manager for nuclear applications development at Gulf Research and Development Company, Pittsburgh.

New Hampshire Assumes Authority

New Hampshire, on May 16, 1966, became the twelfth state to assume part of AEC's regulatory authority over the uses of radioactive materials in the state.

The New Hampshire State Radiation Control Agency of the Department of Health and Welfare, Division of Public Health Services, will conduct the state's radiation control program. The governor's radiation advisory committee will recommend programs and policies to the Agency.

Transfer of authority includes licensing, rule making, and enforcement over the uses of radioisotopes, the source materials uranium and thorium, and small quantities of fissionable materials.

AEC has similar agreements with Arkansas, California, Florida, Kansas, Kentucky, Mississippi, North Carolina, New York, Oregon, Tennessee, and Texas, in addition to New Hampshire.

AEC Exempts Use of ^{3}H in Certain Products

The AEC is amending its regulations to exempt from licensing the possession and use of tritium in a number of consumer products.

The amendments to Commission regulations Part 30 and Part 32 exempt the use of tritium contained in luminous thermostat dials and pointers, automobile shift quadrants, and marine compasses. The manufacture or import of

these items will require a license from the AEC, as is the case with timepieces, automobile-lock illuminators, and precision balances.

The Commission's action results from a petition from Canrad Precision Industries, Inc., New York City, for the exemption. The Commission has determined that the use of tritium in the specified products as authorized will not endanger health and safety.

The amendments, as published for public comment in proposed form on Sept. 17, 1965, also included radio dials and pointers. The Commission has deferred action on these items. The amendments were effective 30 days after publication in the *Federal Register* on Apr. 2, 1966.

AEC Approves First ^{60}Co Production in Michigan Nuclear Power Reactor

AEC approved a recent request by Consumers Power Company of Michigan to produce ^{60}Co in the company's Big Rock Point Nuclear Power Plant, Charlevoix County, Michigan.

Production of the cobalt will be incidental to continued operation of the plant as a power-producing reactor. Full-power operation of the boiling-water reactor results in the production of approximately 72,800 kw(e) net.

This is the first use of a central-station nuclear power plant for the large-scale commercial production of ^{60}Co.

The first cobalt will be inserted into the reactor during the current refueling. AEC also has approved the use of Zircaloy-clad fuel elements in the Big Rock Point reactor, replacing the stainless-steel-clad fuel in the original core.

Polonium SNAP-29 Generator Planned for Short-Lived Earth Orbital Missions

AEC and Martin-Marietta Corporation, Baltimore, are negotiating a contract for development of a nuclear power source for use in short-lived earth orbital missions.

The SNAP-29 generator, fueled with ^{210}Po, will produce 400 watts of power and will weigh about 400 lb. Design studies for the generator were conducted last year. Estimated overall cost of the project is greater than $10 million.

It is expected that such polonium-fueled power systems will eventually be required for Department of Defense and National Aeronautics and Space Administration (NASA) manned and unmanned space programs.

The power level of the polonium generator would be about 10 times greater than any space radioisotope auxiliary power system now being developed by the AEC. Two 30-watt generators are planned for use in a NASA Nimbus weather satellite, and a 50-watt generator will be used in Apollo instrument packages to be left on the moon by astronauts.

Availability of Isotopes and Services

Stable Isotopes Added to ORNL Inventory

Recent additions to the Oak Ridge National Laboratory (ORNL) stable-isotopes inventory include the following:

Isotope	Purity, %	Amount available, g
^{49}Ti	76	10
^{57}Fe	91	28
^{58}Fe	82	3.5
^{67}Zn	92	2.5
^{130}Te	99.5	200

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